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**CHARACTERIZATION OF RADIOACTIVITY IN THE ENVIRONMENT**

**BY**

**ROBERT ANGELO BORRELLI**

**A THESIS**

**SUBMITTED TO THE FACULTY**

**OF THE**

**WORCESTER POLYTECHNIC INSTITUTE**

**IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE**

**DEGREE OF MASTER SCIENCE**

**IN**

**ENVIRONMENTAL ENGINEERING**

**APPROVED: 04 MAY 1999**

## **ABSTRACT**

Ionizing radiation is produced as the result of the decay of an unstable nucleus. The standard measure of radioactivity is quantified according to the rate of disintegration of the unstable nucleus. This method of quantification does not incorporate the total amount of ionizing radiation that is associated with each disintegration of the radionuclide. The ionizing radiation that is produced as a result of decay is specific to a given radionuclide. A radionuclide can be conceptualized as a source of ionizing radiation. Disintegration of the unstable nucleus will therefore result in the continual release of ionizing radiation throughout the fixed existence of the radionuclide.

This thesis will present a reasonable and practical adjustment to the current mechanism regarding the quantification of radionuclides. This adjustment will provide a basis to which the specific decay attributes of radionuclides can be normalized. This Such a normalization will allow for direct comparisons among important inventories of radionuclides. This adjustment will be used to formulate a characterization of common radionuclides that exist in the environment. Such a characterization can provide a control inventory of ionizing radiation to which more specific systems of radionuclides can be compared.

# TABLE OF CONTENTS

<b>LIST OF FIGURES</b>	<b>IV</b>
<b>LIST OF TABLES</b>	<b>V</b>
<b>1.00. BACKGROUND</b>	<b>1</b>
1.10. IONIZING RADIATION	1
1.20. TYPES OF RADIATION	3
1.30. HALF LIFE	6
1.40. SUMMARY	10
<b>2.00. NORMALIZATION</b>	<b>11</b>
2.10. INTRODUCTION	11
2.20. TOTAL ENERGY RELEASE	12
2.30. SUMMARY	21
<b>3.00. ENVIRONMENTAL CHARACTERIZATION</b>	<b>22</b>
3.10. INTRODUCTION	22
3.20. COSMIC SOURCES OF RADIATION	24
3.30. TERRESTRIAL SOURCES OF RADIATION	27
3.40. AREAS OF HIGH RADIOACTIVITY	44
3.50. SUMMARY	47
<b>4.00. APPLICATION</b>	<b>48</b>
4.10. INTRODUCTION	48
4.20. AIRBORNE RELEASES	49
4.30. NEW JERSEY AQUIFER SYSTEM	53
4.40. HIGH LEVEL RADIOACTIVE WASTE	56
4.50. SUMMARY	61
<b>5.00. BIBLIOGRAPHY</b>	<b>62</b>

# LIST OF FIGURES

FIGURE 1.31: THEORETICAL RADIOACTIVE DECAY-----	7
FIGURE 2.21: PARAMETERS GOVERNING THE AREA BOUNDED BY THE DECAY CURVE-----	12
FIGURE 2.22: CUMULATIVE ION PAIR PRODUCTION OF $^{137}\text{Cs}$ AND $^{131}\text{I}$ -----	15
FIGURE 2.23: CUMULATIVE ION PAIR PRODUCTION OF $^{60}\text{Co}$ AND $^3\text{H}$ -----	17
FIGURE 2.24: $^{222}\text{Rn}$ VS DECAY CHAIN -----	18

# LIST OF TABLES

TABLE 2.22.1: ENERGY RELEASE AND ION PAIR PRODUCTION OF $^{137}\text{Cs}$ AND $^{131}\text{I}$ AT 50 YEARS-----	16
TABLE 2.23.1: ENERGY RELEASE ION PAIR PRODUCTION OF $^{60}\text{Co}$ AND $^3\text{H}$ AT 50 YEARS-----	17
TABLE 2.24.1: $^{222}\text{Rn}$ AND THE $^{222}\text{Rn}$ DECAY CHAIN ENERGY RELEASE AND ION PAIR PRODUCTION AT 50 YEARS-----	19
TABLE 2.25: THE $^{222}\text{Rn}$ DECAY CHAIN-----	20
TABLE 3.11: ANNUAL RADIATION EXPOSURE FRACTIONS-----	23
TABLE 3.21: RELEASES OF IONIZING RADIATION AND ION PAIR PRODUCTION OF $^{14}\text{C}$ IN BIOTA-----	26
TABLE 3.22: RELEASES OF IONIZING RADIATION AND ION PAIR PRODUCTION OF $^3\text{H}$ IN THE ENVIRONMENT-----	26
TABLE 3.33: CONCENTRATIONS OF NATURAL RADIONUCLIDES IN SOIL-----	29
TABLE 3.33.1: RELEASES OF IONIZING RADIATION FROM NATURAL RADIONUCLIDES IN SOIL AT 50 YEARS-----	29
TABLE 3.34: RELEASES OF IONIZING ENERGY IN TYPICAL ROCK AT 50 YEARS-----	31
TABLE 3.35: CONCENTRATIONS OF $^{226}\text{Ra}$ IN VARIOUS TYPES OF ROCK-----	34
TABLE 3.35.1: RELEASES OF IONIZING RADIATION IN VARIOUS TYPES OF ROCK AT 50 YEARS-----	34
TABLE 3.36: RELEASES IONIZING RADIATION IN THE BODY AT 50 YEARS-----	35
TABLE 3.37: RELEASES OF IONIZING RADIATION TYPICAL ROCK AT 50 YEARS-----	36
TABLE 3.38: RELEASES FROM RADON IN OUTDOOR AIR AT 50 YEARS-----	38
TABLE 3.39: RELEASES OF IONIZING RADIATION INTO POTABLE WATER SUPPLIES AT 50 YEARS-----	39
TABLE 3.310: RELEASES OF IONIZING RADIATION TO INDOOR AIR AT 50 YEARS-----	41
TABLE 3.311: RELEASES OF IONIZING RADIATION FROM $^{210}\text{Pb}$ IN RAINWATER AND IN THE STANDARD DIET AT 50 YEARS-----	42
TABLE 3.312: RELEASES OF IONIZING RADIATION IN THE BODY FROM $^{210}\text{Pb}$ AND $^{210}\text{Po}$ AT 50 YEARS-----	42
TABLE 3.313: RELEASES OF IONIZING RADIATION OF TYPICAL ROCK AT 50 YEARS-----	43
TABLE 3.42: RELEASES IONIZING RADIATION FROM $^{226}\text{Ra}$ AND $^{222}\text{Rn}$ IN HOT SPRINGS AT 50 YEARS-----	44
TABLE 3.43: RELEASES OF IONIZING RADIATION FROM DIETARY INTAKES IN REGIONS OF BRAZIL AT 50 YEARS-----	45
TABLE 3.44: RELEASES OF IONIZING RADIATION FROM DIETARY INTAKES IN REGIONS OF INDIA AT 50 YEARS-----	46
TABLE 4.21: ATMOSPHERIC RELEASES OF IONIZING RADIATION FROM COAL POWER PLANTS AT 50 YEARS-----	51
TABLE 4.22: ATMOSPHERIC RELEASES OF IONIZING RADIATION FROM A NUCLEAR POWER PLANT AT 50 YEARS-----	51
TABLE 4.31: RELEASES OF IONIZING RADIATION IN KIRKWOOD-COHANSEY AQUIFER SYSTEM AT 50 YEARS-----	53
TABLE 4.24: RELEASES OF IONIZING RADIATION FROM THERMAL SPRINGS AT 50 YEARS-----	55
TABLE 4.51: COMMITTED RELEASES OF IONIZING RADIATION FROM RADIONUCLIDES IN SPENT NUCLEAR FUEL-----	58
TABLE 4.53: RELEASES OF RADIONUCLIDES FROM THE SATURATED ZONE AT 300000 YEARS-----	59
TABLE 4.53.1: EQUIVALENT BACKGROUND RADIATION BASED ON FRACTIONS OF THE GROUNDWATER SYSTEM-----	60

## 1.00. Background

### 1.10. Ionizing Radiation

Radiation is a form of energy, such as light or heat. Radiation is characterized according to the frequencies of these wavelengths. Infrared waves, microwaves, and radio waves occur at the lowest frequencies. Ionizing radiation occurs at the highest frequencies. The process of transforming a neutral atom or molecule into an electrically charged component is known as ionization. Ions are either positively charged or negatively charged, depending upon the number of protons and electrons present in the atom.

Ionizing radiation is produced as the result of the disintegration of an unstable isotope. These isotopes can decay in several ways. The ratio of neutrons to protons in a stable, light atom is approximately unity. This ratio can increase to about 1.5 in heavier elements.<sup>1</sup> Unstable nuclei exist because of an imbalance in this ratio. The interaction of this ionizing radiation with specific biological material will result in the production of ion pairs. An ion pair is essentially an ionized atom together with its ejected electron. Therefore, ionizing radiation will affect the fundamental structures of biological materials through the production of these ion pairs.

Several different species of radionuclides will produce ionizing radiation. Heavy elements, which include radionuclides such as <sup>238</sup>U and <sup>232</sup>Th, are found in all common types of rock and soil. Furthermore, these radionuclides disintegrate into other nuclides that are also radioactive, thus forming a decay chain. Therefore, not only is the release of ionizing radiation cumulative over the existence of the lead radionuclide in a decay chain, but additive throughout all the energy releases from the progeny in the decay chain as well. Products formed in a nuclear reactor as the result of nuclear fission are radioactive. Other radioactive isotopes can be produced by particle bombardment processes, such as neutron activation.

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<sup>1</sup> Brookhaven National Laboratory, "Introduction to Tritium and Radioactivity," 05-May-1997, [<http://www.hfbr.bnl.gov/hfbrweb/hdb1079a.html>].

### **1.11. Radioactive Decay**

Radionuclides are quantified according a rate of disintegration. This rate is defined by the Curie and the Becquerel. One Curie is defined as  $1 \text{ Ci} = 3.7 \times 10^{10}$  disintegrations per second, and one Becquerel is defined as  $1 \text{ Bq} = 1$  disintegration per second. These units, however, incorporate neither the total amount of ionizing energy associated with each disintegration, nor the total existence of the radionuclide. Singular usage of the Curie or the Becquerel also does not fully reflect the cumulative quantity of ionizing radiation that will be imparted to a specific biological material.

The energy accompanying each radioactive disintegration is specific to the given radionuclide, and is dependent upon the amount of source material present. Therefore, the quantity of ionizing radiation that will be released from a radionuclide is specific to each radionuclide. The total quantity of ionizing radiation that will be released is essentially based upon a fixed set of radioactive particles and photons that are ejected during disintegration. Additional factors such as the energy release per each disintegration of the radionuclide, as well as the length of time that the radionuclide will exist, can provide practical adjustments to the sole use of the unit of activity as a quantification device.

### **1.12. Specific Ionization**

The unstable isotope will disintegrate to the stable state through the emission of excess energy in the form of photons, particles, or a combination of both. Each of these particles and photons is ejected from the radioactive source at a certain energy level, specific to the source material. The extent to which ejected particles and photons interact with matter can be described in terms of specific ionization.

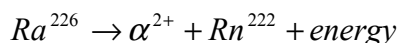
Specific ionization can be defined as the quantity of ion pairs that will be produced, given a specific amount of ionizing radiation that a radionuclide will release. Approximately 34 eV are required to produce one ion pair. The rate of production of ion pairs is energy specific and not dependent upon the type of radioactive particle involved in the interaction. Therefore, a 1 MeV release accompanying the emission of a radioactive particle or a photon will produce approximately 30000 ion pairs for every release. As a result of the continuous release of energy from a radioactive source, ion pairs will be generated continually throughout the existence of the source. A quantity of radioactivity can be measured in terms of the number of ion pairs that the energy will produce in exposed material. This adjusted measure of radioactivity can be applied to the vast and highly variable amounts of sources that exist in nature. Such adjustments can aid in the assessment of the environmental effects of a source defined in terms of the imparted energy to materials, based upon the number ion pairs that the source will produce.



## **1.20. Types of Radiation**

### **1.21. Alpha Particles**

An alpha particle is comprised of two protons and two neutrons. The alpha particle possesses an electrical charge of +2. Alpha particle decay will occur from a nucleus in which the number of neutrons is much higher than the number of protons. Nuclei heavier than lead, such as thorium, plutonium, radon, and uranium decay through alpha emission.



Each radionuclide will emit one or more alpha particles at fixed energy levels. The resulting release of ionizing radiation will be imparted to a given medium only over a small distance. Alpha particles collide very easily with matter and lose energy quickly because of large size and of high electrical charge. Only a few centimeters of air, or the outer layer of skin, will provide a sufficient barrier to alpha particle penetration.

The rate of energy that is imparted to a body from a radioactive particle is defined by the linear energy transfer (LET). The quantity of LET radiation imparted to a medium will increase rapidly with the mass and the charge of the particle.<sup>2</sup> Therefore, alpha particle decay will impart the highest quantities of LET radiation. Particles that decay as high LET radiation have the potential to produce a greater biological effect in an absorbing medium.

### **1.22. Beta Particles**

Beta particles are electrons that have been ejected by the nucleus. Nuclei decay through beta particle emission through the emission of a negative beta particle (negatron) or a positive beta particle (positron). Regardless of the nature of the beta particle, sources that emit this form of decay encompass a wide range of energies. Unlike alpha particles, beta particles are found to be emitted with a continuous energy distribution ranging from zero to a theoretically expected maximum energy. This theoretical value is based upon mass and energy considerations for each specific beta particle transition.<sup>3</sup> The energies listed for radionuclides that decay through the emission of beta particles on common scientific tables are the theoretically maximum energies that could occur. The average energy release from a beta particle is approximately 30% to 40% of the theoretical maximum.<sup>4</sup>

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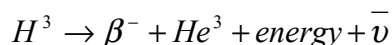
<sup>2</sup> Lamarsh, 89.

<sup>3</sup> Cember, 64.

<sup>4</sup> Cember, 64.

### 1.22.1. Negatrons

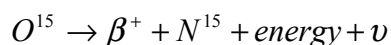
Negatron emission results from a condition in the nucleus in which the neutron to proton ratio is high. During this type of decay, a neutron emits the negatron, and then is converted into a proton. The beta particle possesses an electric charge of -1.



Unlike alpha decay, however, negatron emission occurs over a wide range of radioisotopes. Negatrons have the size and the mass of an electron. As such, they can travel farther and penetrate deeper than alpha particles. Some of the highest energy negatrons can penetrate a few centimeters of water or organic tissue. Common negatron emitting isotopes found in research laboratories today include  $^{32}\text{P}$ , and  $^{35}\text{S}$ . A thin plastic shield (less than 1") will provide a sufficient barrier to the lowest energy negatrons. A thin aluminum shield may be required as a barrier to the highest energy negatrons.

### 1.22.2. Positrons

Positron emission is another form of beta particle decay. Unlike the previous form of beta decay, positron emission results from an excessively high proton to neutron ratio. The proton emits a positron, and then is converted to a neutron. The positron possesses an electric charge of +1.



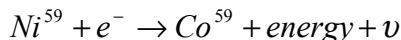
This form of beta particle decay will occur over a wide range of radioisotopes as well. Positrons also have the mass and the size of an electron. As such, the relative penetrations of positrons are essentially identical to negatron penetrations. Positrons, however, are always accompanied by the .511 MeV release of energy that arises as the result of the annihilation of a slow positron with a negative electron. Similar shielding requirements are required for positrons.

### 1.23. Electron Capture

Like positron emission, electron capture can occur when the proton to neutron ratio is excessively high. If a neutron deficient atom is to attain stability by positron emission, then the atom must exceed the weight of the daughter by at least two electron masses.<sup>5</sup> Lacking the energy for positron emission, electron capture will occur.

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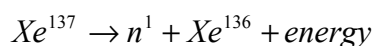
<sup>5</sup> Cember, 68.



In this process, an atomic electron from the K shell interacts with one of the protons in the nucleus. A neutron is subsequently formed. The subsequent vacancy in the electron cloud is then filled by the Auger electron.

### **1.24. Neutrons**

Another form of decay includes neutron emission. This form of decay predominantly occurs as the result of spontaneous fission. The phenomenon of spontaneous fission is the initial component in the creation of a controlled chain reaction in a nuclear reactor core. Many heavy metals, such as those found in the actinide series, are spontaneously fissioning radionuclides. These fission events are accompanied with other forms of decay as well. Fission products formed in a reactor core also emit a neutron as the dominant form of decay. These delayed neutrons are the basis for nuclear reactor operation.



Neutrons possess about one quarter of the mass of an alpha particle. Providing a barrier with which to shield the penetration of neutrons is very difficult because these particles carry no electric charge. Neutrons will pass through an electron cloud and interact directly with the nucleus of another atom. Such interactions and collisions result in subsequent fissions. In addition, other forms of radiation will occur. Collisions with hydrogen nuclei are most effective in stopping or slowing neutrons.<sup>6</sup> Therefore, products with a high hydrogen content, such as water and various plastics, can provide effective barriers to neutron penetration.

### **1.25. Gamma Rays and X Rays**

Unlike the previous types of decay, gamma rays represent energy that is transmitted in a wave. Gamma rays are similar to x rays in that both forms of radiation are photons. Fundamentally, there is no difference between these types of photons. Gamma rays originate in the nucleus, while x rays result from transitions of atomic electrons in the electron cloud. These types of radiation occur at the highest frequencies of the energy spectrum. Gamma rays occur at a higher frequency than x rays.

Nuclei formed as a result of the previous methods of decay usually are left in an excited state after particle emission. Excited nuclei then proceed to decay to the ground state through the emission of gamma rays or x rays. A large majority of nuclei in this excited state decay in an immeasurably short period of time. In some instances, the decay of excited nuclei is delayed.

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<sup>6</sup> Brookhaven National Laboratory, "Introduction to Tritium and Radioactivity," 05-May-1997, [<http://www.hfbr.bnl.gov/hfbrweb/hdb1079a.html>].

Excited nuclei appear to exist in a semistable, or isomeric, state. The subsequent decay, an isomeric transition, occurs as the semistable nucleus emits a gamma ray or an x ray in order to exist at the ground state. Beta particle emission also can take place in the isomeric state.

Because gamma rays and x rays are energy waves, they do not possess a mass or an electrical charge. These photons have the highest ranges of penetration. These forms of radiation can pass through the human body. Highly dense materials, such as lead or steel, provide effective barriers to gamma rays and x rays.

### **1.30. Half Life**

Each radionuclide will decay for a fixed amount of time. This amount of time defines the existence of each radionuclide. Scientists, engineers, and other professionals in nuclear related fields use these fixed existences with which to classify radioisotopes. The time that has elapsed for the activity of a radioisotope to decrease by a factor of two is defined as a half life. The probability per unit time that a radioactive nuclide will decay is a constant and independent of time.<sup>7</sup>

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<sup>7</sup> Lamarsh, 22.

## Theoretical Radioactive Decay

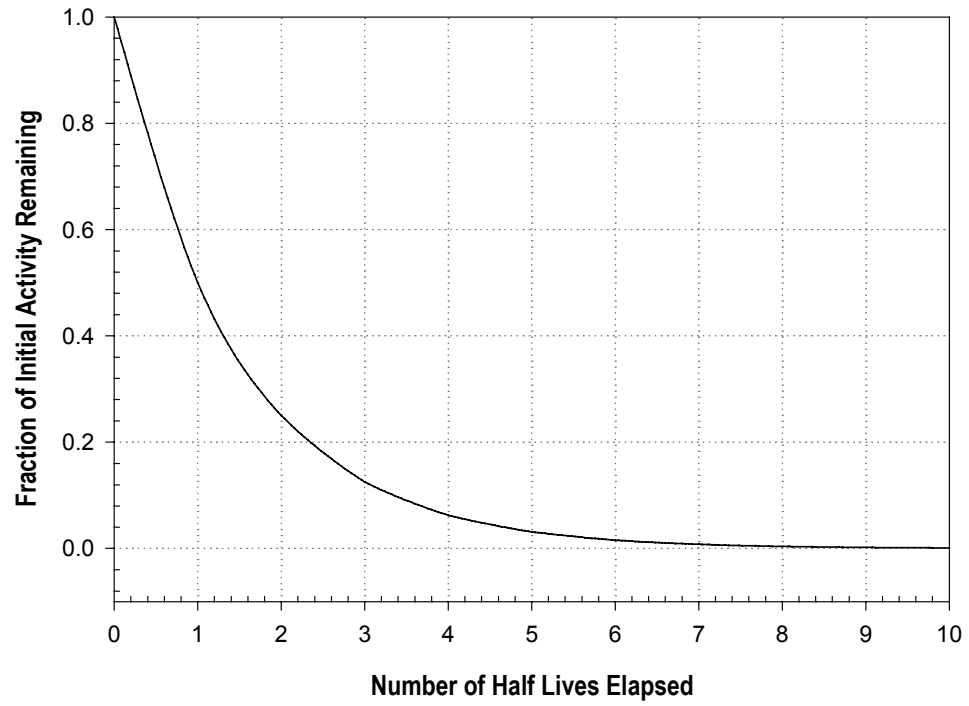


FIGURE 1.31: THEORETICAL RADIOACTIVE DECAY

### 1.31. Derivation

Figure 1.32 shows that the decay of a radioactive isotope follows a first order, exponential model. Assume that at a given time,  $t$ , there are  $N$  existing radioactive atoms of a given radioactive source. The rate of change in the number of radioactive atoms in a given time interval  $dt$  can be expressed as:

$$\frac{dN}{dt} = -\lambda N$$

In this model,  $\lambda$  is defined as the decay constant. Assume that  $N = N_0$  radioactive atoms are present when  $t = 0$ .

This model can be further developed:

$$\frac{dN}{N} = -\lambda dt$$

$$\int_{N_0}^N \frac{dN}{N} = -\lambda \int_0^t dt$$

$$\ln\left(\frac{N}{N_0}\right) = -\lambda t$$

$$N = N_0 e^{-\lambda t}$$

This model represents the number of undecayed nuclei in a radioactive sample for a given time. This model now can be developed further in order to determine the quantity of radioactive atoms of source material that will disintegrate during a given unit of time. Ultimately, this model can be adjusted in order to quantify the amount of ionizing radiation that will be released from a specific radionuclide.

The rate of disintegration, defined as the activity of the sample, then can be expressed as:

$$\lambda \cdot N = \lambda \cdot N_0 e^{-\lambda t}$$

or:

$$\alpha = \alpha_0 e^{-\lambda t}$$

The half life then can be determined as follows:

$$\frac{1}{2} \alpha_0 = \alpha_0 e^{-\lambda t_{1/2}}$$

$$\ln\left(\frac{1}{2}\right) = -\lambda t_{1/2}$$

$$-\ln(2) = -\lambda t_{1/2}$$

$$t_{1/2} = \frac{\ln(2)}{\lambda}$$

$$\lambda = \frac{\ln(2)}{t_{1/2}}$$

$$\alpha = \alpha_0 e^{\left(\frac{-\ln(2)}{t_{1/2}}\right) t}$$

## **1.40. Summary**

Unstable nuclei possess a high imbalance between protons and neutrons. Radioactive decay will occur through the disintegration of the unstable nucleus. The disintegration will result in the emission of several particles, photons, or a combination of both. The emission of these particles and photons from the unstable nucleus will be accompanied by a subsequent release of ionizing energy.

Not all radionuclides decay directly to the stable state. Some of these radionuclides will decay to another nuclide that is itself unstable, thus forming a chain of unstable nuclei. These types of chains are common in nature. The releases of ionizing energy from all subsequent radionuclides in the decay chain must be added to the energy release of the lead radionuclide to incorporate the continuous decay that occurs. The full characterization of the release of ionizing radiation from a radionuclide that leads a decay chain not only is cumulative over the existence of the parent radionuclide, but additive over the existences of each radionuclide in the decay chain as well.

Each radioactive source decays at a fixed rate. Therefore, each radioactive source will exist for a fixed amount of time. A sample of radioactive material can be expressed as a source of particle emissions occurring at specific energies. These factors contribute to a cumulative release of ionizing energy over the fixed existence of the source material. The energies at which a source will disintegrate as well as the rate at which a source will disintegrate is specific to each radioisotope. A model representing the actual adjustment to the quantification of radionuclides from the perspective of the release of ionizing energy, can be developed through an examination of the nature of radioactive decay over time.



## **2.00. Normalization**

### ***2.10. Introduction***

Section 1.00 presented the concept that a specific radionuclide is essentially a source of radioactive particles and photons that will continually release ionizing radiation. Each radionuclide will exist for a fixed amount of time. The singular usage of the unit of activity, as a method with which to quantify radionuclides, neglects factors such as half life and decay energies. A model that incorporates the concept of the radionuclide as a source of ionizing energy can be developed for utilization as an adjustment to the present manner of radionuclide quantification.

### ***2.11. Variables***

The total amount of ionizing radiation that a radionuclide will release is dependent upon a combination of the quantity of radioactive material that is available for decay, the ionizing radiation energy that is associated with each disintegration of the radionuclide, the half life of the radionuclide, and the given framework of time that is available for analysis. The quantification of a radionuclide can be adjusted to account for the cumulative release of ionizing energy, based upon these factors.

## 2.20. Total Energy Release

### 2.21. Derivation

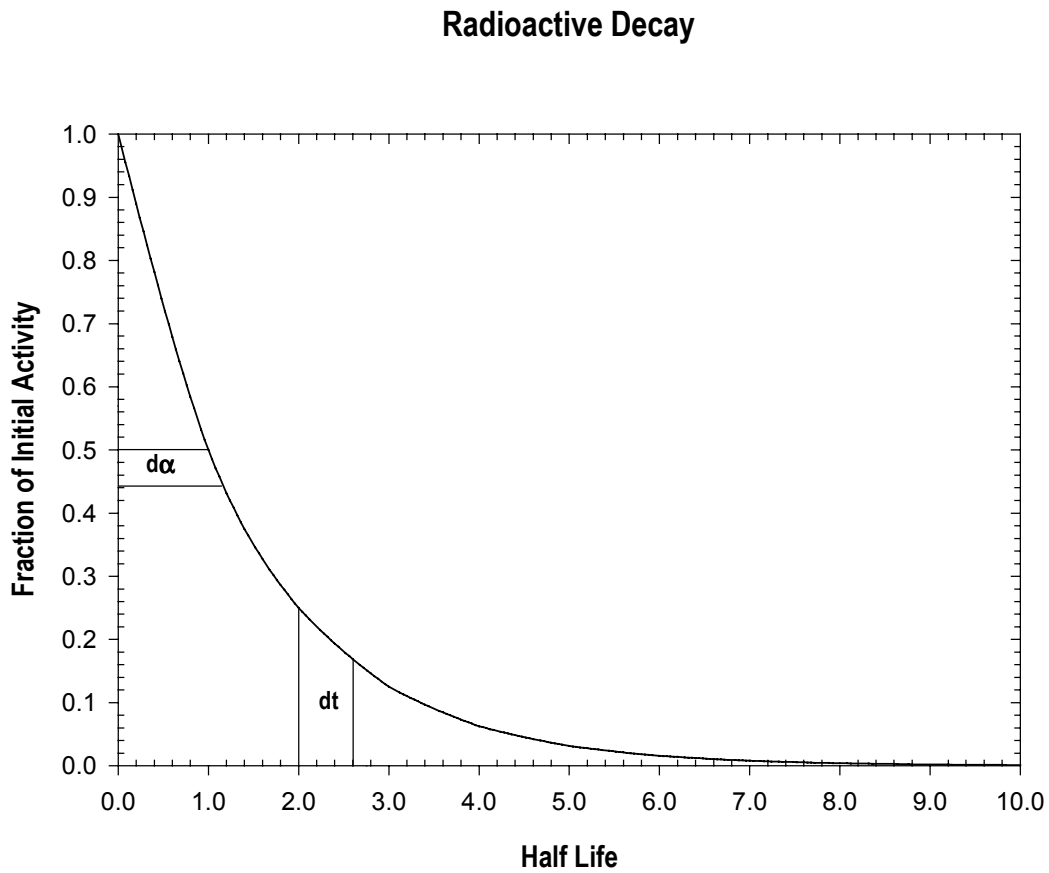


FIGURE 2.21: PARAMETERS GOVERNING THE AREA BOUNDED BY THE DECAY CURVE

Figure 2.21 represents the fractional decay of a radionuclide over a given time period. The area that is bounded by this decay curve, however, essentially describes the quantity of ionizing radiation that the radionuclide will release. The area bounded by the decay curve is dependent upon the rate of change of the activity of the radionuclide per unit time.

The total release of ionizing radiation from a radionuclide can be expressed in the most general sense as:

$$E = \int dA$$

Such that:

$$E = \int dA = \int \alpha dt$$

Where:

$$\alpha = \alpha_0 e^{-\lambda t}$$

In this equation activity is assumed to be in Curies, as derived in Section 1.31.

The total release of ionizing energy per unit time,  $dt$ , such that  $dt = t_f - t_0$ , then can be expressed as:

$$E = \int_{t_0}^{t_f} \alpha dt = \int_{t_0}^{t_f} \alpha_0 e^{-\lambda t} dt$$

Evaluating the integral:

$$E = \int_{t_0}^{t_f} \alpha dt = \frac{-\alpha_0}{\lambda} e^{-\lambda t} \Big|_{t_0}^{t_f}$$

and:

$$E = \int_{t_0}^{t_f} \alpha dt = \frac{-\alpha_0}{\lambda} e^{-\lambda t} \Big|_{t_0}^{t_f}$$

Therefore:

$$E = \frac{\alpha_0}{\lambda} [e^{-\lambda t_0} - e^{-\lambda t_f}]$$

Allowing  $t_0 = 0$  and  $t_f = t$ :

$$E = \frac{\alpha_0}{\lambda} [1 - e^{-\lambda t}]$$

The model now must be adjusted to reflect an energy release rate. The development of the energy release rate for the model results in the expression of the radionuclide in terms of a disintegration rate, as well as the summation of the ionizing energy associated with each disintegration of the radionuclide, in which  $n$  is the total number of particles or of photons ejected from the unstable nucleus.

Define a new constant:

$$\Gamma \equiv \frac{MeV}{s}$$

Such that:

$$\Gamma \equiv \alpha_0 \cdot 3.7 \times 10^{10} \frac{dps}{Ci} \cdot \sum_{i=1}^n \frac{[MeV \cdot f]_i}{d}$$

Therefore:

$$E = \frac{\Gamma}{\lambda} [1 - e^{-\lambda t}]$$

This model represents the total quantity of ionizing radiation that a radionuclide will continually release over a given time period. The model incorporates the half life of the radionuclide and the energy associated with each disintegration. These characteristics are unique to each specific radionuclide.

## 2.22. Examples

The central focus in the development of this model is that each radionuclide exhibits characteristics that are unique to each specific radionuclide. These characteristics can be used to determine the quantity of ionizing radiation that each radionuclide will release. These quantities of ionizing radiation can be used in order to aid in the determination and in the assessment of both the biological effects and the environmental effects of radionuclides within a given system. The usage of the unit of activity will only result in the quantification of the decay events that will occur. Quantification of radionuclides from the perspective of a total release of ionizing energy

normalizes those factors that are unique to each specific radionuclide. This normalization thus allows for a basis to which different radionuclides can be directly compared.

### 2.22.1 Long Lived Radionuclides vs Short Lived Radionuclides

Two common radioisotopes found in nuclear power plants are  $^{131}\text{I}$  and  $^{137}\text{Cs}$ . These radionuclides are found in a nuclear reactor core as fission products. While both radionuclides decay through the emission of a negatron, the half life of  $^{137}\text{Cs}$  is greater than that of  $^{131}\text{I}$  by a factor of approximately 1000. Therefore, starting with the same amount of activity,  $^{131}\text{I}$  will have decayed fully before much of the  $^{137}\text{Cs}$  decays at all.

#### Long Lived vs Short Lived

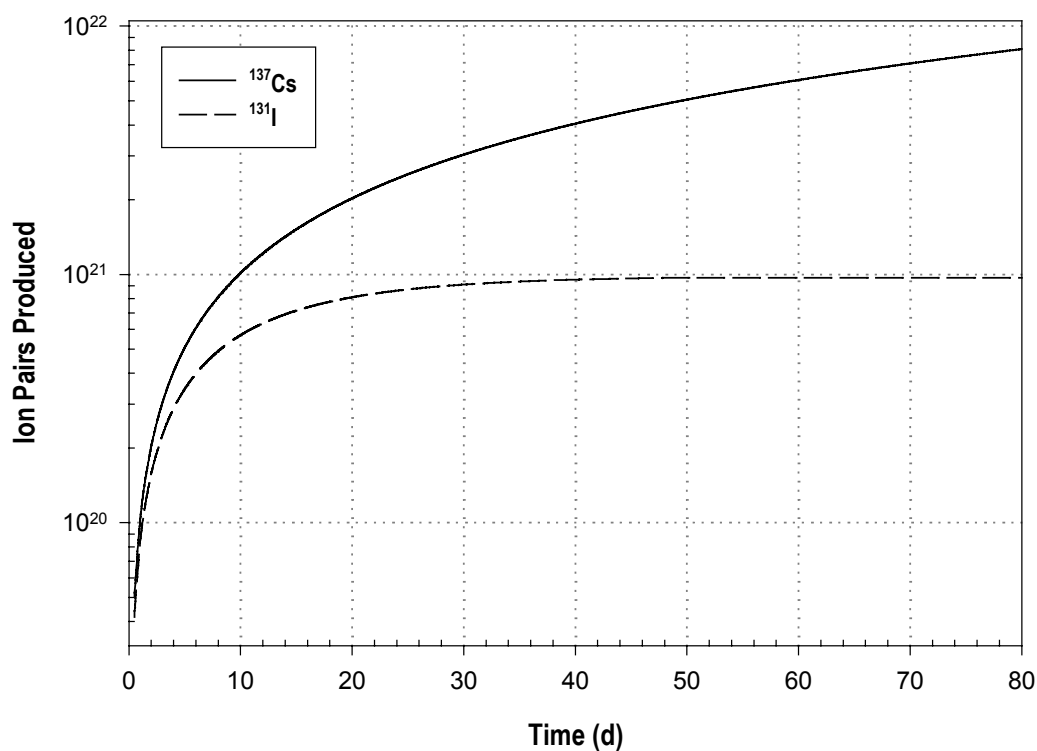


FIGURE 2.22: CUMULATIVE ION PAIR PRODUCTION OF  $^{137}\text{Cs}$  AND  $^{131}\text{I}$

Radionuclide	Activity (Ci)	Energy Release (MeV)	Ion Pairs Produced
$^{137}\text{Cs}$	1.	$275. \times 10^{15}$	$8. \times 10^{21}$
$^{131}\text{I}$	1.	$33. \times 10^{15}$	$983. \times 10^{18}$

TABLE 2.22.1: ENERGY RELEASE AND ION PAIR PRODUCTION OF  $^{137}\text{Cs}$  AND  $^{131}\text{I}$  AT 50 YEARS

Although both of these radionuclides are fission products and decay at similar energy levels, Figure 2.22 clearly shows that the usage of the unit of activity as a basis for comparison neglects the other characteristics that are unique to these radionuclides. Essentially, 1 Ci of  $^{137}\text{Cs}$  is not equivalent to 1 Ci of  $^{131}\text{I}$ . Furthermore, the addition of 1 Ci of  $^{137}\text{Cs}$  with 1 Ci of  $^{131}\text{I}$ , to characterize the quantity of fission products in the reactor core, for example, does not result in a total of 2 Ci of radioactivity. The addition of 1 Ci of  $^{137}\text{Cs}$  with 1 Ci of  $^{131}\text{I}$  will result, however, in a total committed energy release of approximately  $308. \times 10^{15}$  MeV of ionizing radiation.

### 2.22.2 High Energy Radionuclides vs Low Energy Radionuclides

Tritium is produced naturally as a result of interactions between gases in the upper atmosphere and cosmic rays. This radionuclide is also produced as a fission product in a nuclear reactor core.  $^{60}\text{Co}$  can be produced artificially. This radionuclide is used for thickness gauges, for gamma radiography, and for food irradiation.  $^{60}\text{Co}$  also is produced as a neutron activation product in various pieces of equipment that are found in a nuclear reactor core.

Tritium decays with approximately an 18 keV negatron, while  $^{60}\text{Co}$  decays with approximately a 300 keV negatron. In addition,  $^{60}\text{Co}$  emits two gamma rays during decay, while tritium does not emit any gamma rays. Singular usage of the Curie with which to quantify these radionuclides essentially neglects these factors. By expressing these radionuclides as sources of ionizing energy, a more accurate characterization of the manner in which tritium and  $^{60}\text{Co}$  decay can be provided.

## High Energy vs Low Energy

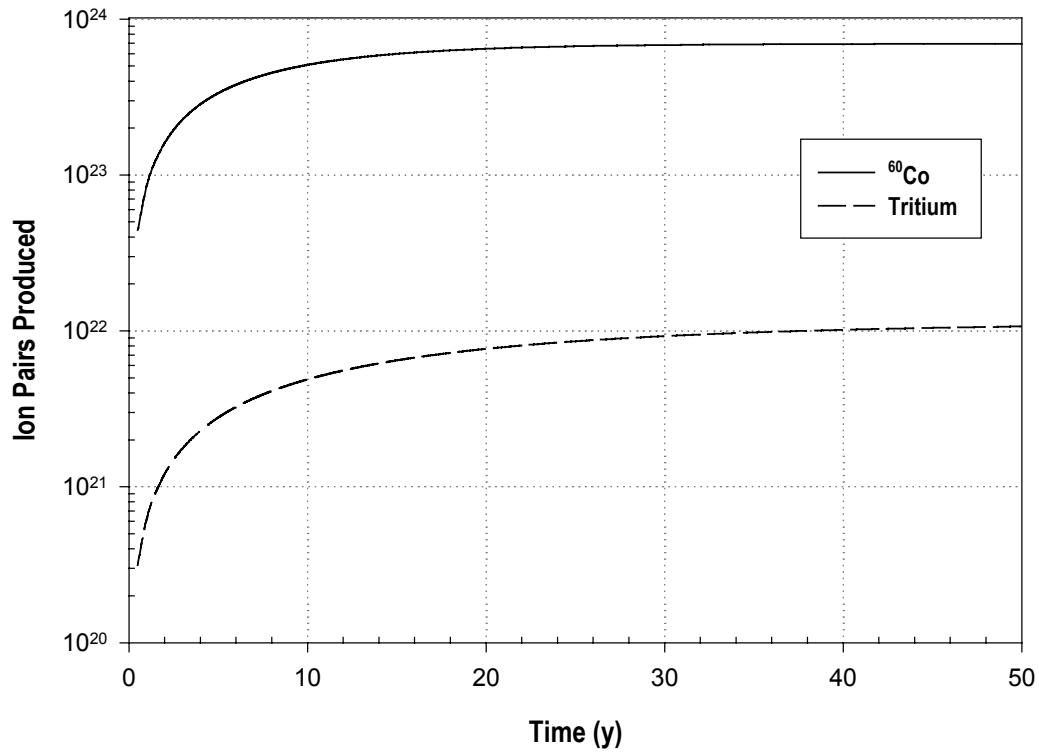


FIGURE 2.23: CUMULATIVE ION PAIR PRODUCTION OF  $^{60}\text{Co}$  AND  $^3\text{H}$

Radionuclide	Activity (Ci)	Energy Release (MeV)	Ion Pairs Produced
$^{60}\text{Co}$	1.	$25. \times 10^{18}$	$736. \times 10^{21}$
$^3\text{H}$	1.	$363. \times 10^{15}$	$11. \times 10^{21}$

TABLE 2.23.1: ENERGY RELEASE ION PAIR PRODUCTION OF  $^{60}\text{Co}$  AND  $^3\text{H}$  AT 50 YEARS

### 2.22.3. Decay Chains

Some radionuclides exist as constituents of a decay chain, such as  $^{222}\text{Rn}$ . This radionuclide is significant to study because the  $^{222}\text{Rn}$  decay chain accounts for the greatest fraction of natural exposures to ionizing radiation. The most significant mechanism of exposure to the  $^{222}\text{Rn}$  decay chain is the inhalation of the decay products of the parent radionuclide. Over the existence of a radionuclide such as  $^{222}\text{Rn}$ , that leads a decay chain, the total amount of ionizing radiation that will provide the basis of exposures to humans not only are the energy releases attributable to  $^{222}\text{Rn}$  itself, but the energy releases of each daughter product in this decay chain as well.

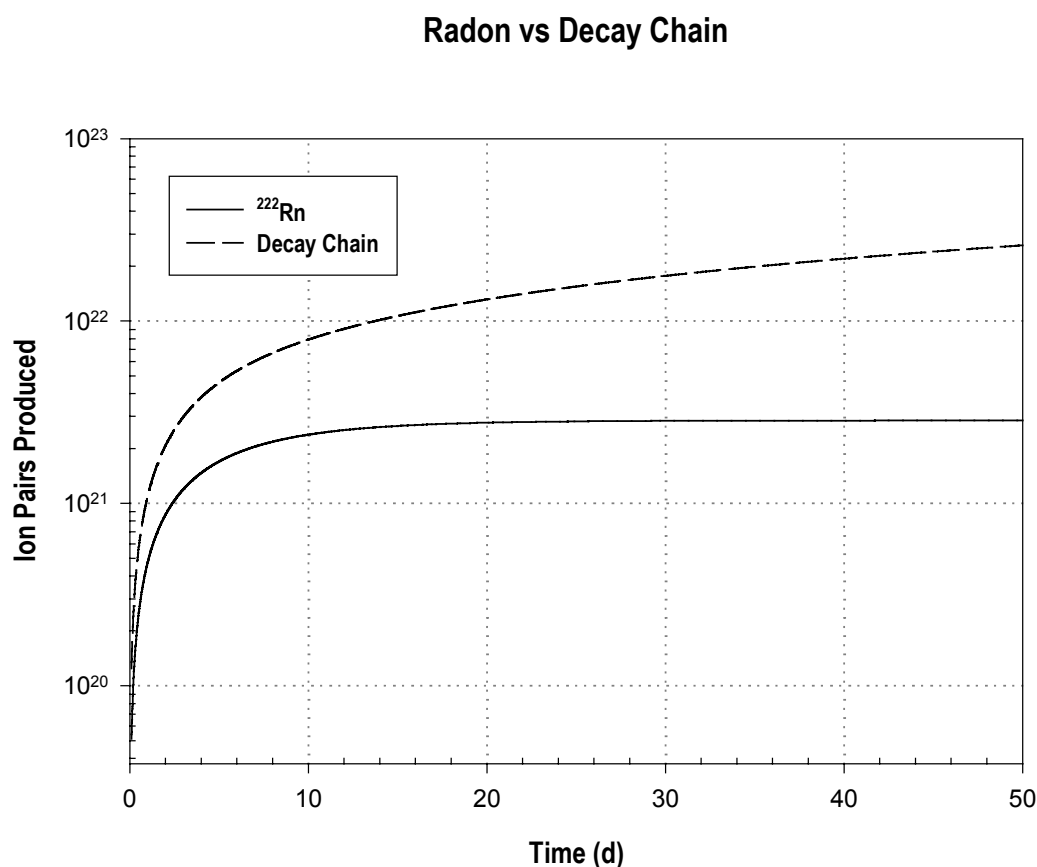


FIGURE 2.24:  $^{222}\text{Rn}$  VS DECAY CHAIN



Radionuclide	Activity (Ci)	Energy Release (MeV)	Ion Pairs Produced
$^{222}\text{Rn}$	1.	$97. \times 10^{15}$	$3. \times 10^{21}$
Full Decay Chain	1.	$884. \times 10^{15}$	$26. \times 10^{21}$

**TABLE 2.24.1:  $^{222}\text{Rn}$  AND THE  $^{222}\text{Rn}$  DECAY CHAIN ENERGY RELEASE AND ION PAIR PRODUCTION AT 50 YEARS**

$^{222}\text{Rn}$  decays with a half life of approximately 4 days. Therefore, after approximately 40 days have elapsed the rate of release of ionizing radiation attributable to  $^{222}\text{Rn}$  itself will become negligible, as indicated by the leveling of the  $^{222}\text{Rn}$  energy release curve in Figure 2.24. The releases of ionizing radiation attributable to the full decay chain are dominated by  $^{210}\text{Pb}$ , which decays with a half life of approximately 22 years, and  $^{210}\text{Po}$ , which decays with a half life of approximately 138 days. The remaining radionuclides in this chain decay with half lives in the minutes and in the seconds. Radionuclides such as  $^{222}\text{Rn}$ , which lead decay chains, cannot be singularly represented through the activity and the characteristics of only the parent radionuclide.

Radionuclide	Half-Life	Half-Life (years)	Decay Mode	Fraction (Intensity)	Energy (MeV)
Radon-222	3.8235 days	1.0468E-02	Alpha	0.999200	5.489520
Radon-222			Alpha	0.000780	4.987000
Radon-222			Gamma Ray	0.000760	0.510000
Polonium-218	3.10 minutes	5.8940E-06	Alpha	0.999789	6.002400
Polonium-218			Negatron	0.000200	0.264000
Astatine-218	1.6 seconds	5.0701E-08	Alpha	0.035694	6.746000
Astatine-218			Alpha	0.899100	6.694000
Astatine-218			Alpha	0.069360	6.653000
Astatine-218			Negatron	0.001000	2.883000
Lead-214	26.8 minutes	5.0954E-05	Negatron	1.000000	0.670000
Lead-214			Gamma Ray	0.075000	0.241981
Lead-214			Gamma Ray	0.185000	0.295213
Lead-214			Gamma Ray	0.358000	0.351921
Bismuth-214	19.9 minutes	3.7836E-05	Alpha	0.000082	5.516000
Bismuth-214			Alpha	0.001132	5.452000
Bismuth-214			Negatron	0.999790	3.272000
Bismuth-214			Gamma Ray	0.447906	0.609312
Bismuth-214			Gamma Ray	0.147969	1.120287
Bismuth-214			Gamma Ray	0.028794	1.729595
Polonium-214	164.3 micro	5.2064E-12	Alpha	0.999895	7.686820
Polonium-214			Alpha	0.000104	6.902100
Polonium-214			Gamma Ray	0.000104	0.799700
Thallium-210	1.30 minutes	2.4717E-06	Negatron	1.000000	2.300000
Thallium-210			Gamma Ray	0.791680	0.296000
Thallium-210			Gamma Ray	0.989600	0.799600
Thallium-210			Gamma Ray	0.118752	1.070000
Thallium-210			Gamma Ray	0.168232	1.210000
Thallium-210			Gamma Ray	0.207816	1.316000
Lead-210	22.3 years	2.2300E+01	Negatron	1.000000	0.061522
Lead-210			Gamma Ray	0.042500	0.046539
Bismuth-210	5.013 days	1.3725E-02	Alpha	0.000001	4.694000
Bismuth-210			Alpha	0.000001	4.656000
Bismuth-210			Negatron	1.000000	1.162700
Polonium-210	138.376 days	3.7885E-01	Alpha	1.000000	5.304380
Polonium-210			Gamma Ray	0.000012	0.803100
Thallium-206	4.199 minutes	7.9835E-06	Negatron	1.000000	1.533200
Thallium-206			Gamma Ray	0.001000	1.165000

TABLE 2.25: THE <sup>222</sup>Rn DECAY CHAIN

### **2.30. Summary**

A radionuclide can essentially be characterized as a source of subatomic particles and of photons that each will emit a specific quantity of ionizing energy. This release of ionizing energy is cumulative over the entire existence of the radionuclide. Several factors that are unique to each specific radionuclide will affect the overall determination of the quantity of ionizing radiation that each specific radionuclide will release. Such factors include the quantity of source material that is available for decay, the energy associated with each disintegration of the source, the half life of the source, and the overall time frame of an analysis. The strict usage of activity does not incorporate these factors into the quantification of a radionuclide. Furthermore, the representations of radionuclides that lead decay chains that are expressed singularly in terms of the activity of the parent radionuclide, do not incorporate and do not reflect the cumulative releases of ionizing radiation that are associated with the daughter products of the decay chain. Therefore, representations and comparisons of radionuclides based solely on the unit of activity do not accurately and completely characterize the manner in which each specific radionuclide exists with relation to other radionuclides within a given system or a given environment.

A practical adjustment to the quantification of radionuclides has been developed in order to enable more direct comparisons among important inventories of radionuclides. Furthermore, more accurate assessments can be formulated regarding these important inventories of radionuclides, as cumulative releases of ionizing radiation can now be added together in order to develop an overall quantity of ionizing radiation that will be characteristic of the inventory as a whole. Therefore, characterizations of systems of radioactivity based on cumulative releases of ionizing energy can provide an aid to existing characterizations with regard to significant exposures to both the global population and local populations.

## 3.00. Environmental Characterization

### 3.10. Introduction

Section 2.00 provided the derivation of a model that has incorporated the various factors that are unique to each specific radionuclide into a solution that will express each radionuclide as a single source of ionizing radiation. By adjusting the quantification of radionuclides from the sole use of the unit of activity, to an overall release of ionizing energy, differences among radionuclides, such as half lives and decay energies have been normalized. This normalization can aid in the formulation of direct comparisons between significant systems of radionuclides, as well as further developments in existing characterizations of such systems.

Natural sources of radiation are the concentrations of radionuclides that represent ambient conditions present in the environment that are in no way influenced by human activity.<sup>8</sup> Everyone on the planet is exposed to some background level of ionizing radiation. External exposures will occur as a result of irradiation, and internal exposures will occur as a result of inhalation. Furthermore, because of the extensive quantities of radionuclides in soil, and in vegetation, exposures also will occur as a result of ingestion.

Sources of radiation originating in nature can be divided into four general categories. Cosmic rays originate from outer space. This type of radioactivity consists of very high energy particles and photons. Cosmogenic radionuclides are formed as a result of the interaction of cosmic rays with nuclides in the upper atmosphere. Primordial radionuclides have half lives comparable to the age of the earth. These radionuclides are found in all common rock and soil. Due to the variety of sources of ionizing radiation found in the environment, there is a considerable variation to exposures from these sources throughout the world.

Natural sources of radiation provide an overwhelming contribution to annual exposures received by the population of the world. Therefore, an extensive characterization of sources of radioactivity can serve as a basis to which the effects of other, more specific inventories can be compared. Singular usage of the activity of a source with which to quantify these inventories does not allow for a complete and an accurate characterization. Therefore, employment of the adjusted method of quantification to these radionuclides will provide the opportunity for a more effective analysis of the effects of ionizing radiation with respect to significant human exposures.

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<sup>8</sup> Phil Egidi, "Background," 26-January-1998, [<http://www.normis.com/bkgnd.htm>].

Sources	Per Cent of Total
<b>Natural</b>	
Cosmic	8%
Internal	11%
Radon	55%
Terrestrial	8%
<b>Total</b>	<b>82%</b>
<b>Artificial</b>	
Consumer Products	3%
Fallout	less than .03%
Medical X-Rays	11%
Miscellaneous	less than .03%
Nuclear Fuel Cycle	less than .03%
Nuclear Medicine	4%
Occupational	less than .3%
<b>Total</b>	<b>18%</b>
<b>Total</b>	<b>100%</b>

**TABLE 3.11: ANNUAL RADIATION EXPOSURE FRACTIONS<sup>9</sup>**

<sup>9</sup> Health Physics Society of the University of Michigan, "Radiation and Us," April-1997, [<http://www.sph.umich.edu/group/eih/UMSCHPS/radrus.htm>].

## **3.20. Cosmic Sources of Radiation**

### **3.21. Cosmic Radioactivity**

Cosmic radiation exists throughout the universe, originating from stellar flares, supernovas, and pulsars. Cosmic radiation consists of high energy particles, such as protons, high energy electrons, alpha particles, and light atomic nuclei. Cosmic radiation is highly penetrating, with a mean energy of approximately  $10^4$  MeV.<sup>10</sup> Some of this cosmic radiation originates from the sun. Solar particles are generated continuously, but the solar contribution to primary cosmic radiation only becomes significant following flares attributed to increased, cyclical sunspot activity. A majority of solar particles lacks sufficient energy to penetrate the planetary magnetic field.

Interactions of cosmic rays with gases in the upper atmosphere will produce such subatomic particles as electrons, protons, neutrons, and muons. Most of these particles will dissipate before entry into the lower atmosphere because of energy losses due to nuclear collisions. Muons, however, will still penetrate to sea level because these particles possess a small cross section for interaction. Most muons occur in the energy range of 200 MeV to 20000 MeV with a median energy of 2000 MeV.<sup>11</sup> Muons contribute to a majority of the cosmic ray flux at sea level.

The planetary magnetic field traps some forms of highly energetic cosmic radiation. The magnetic field deflects some cosmic particles toward the polar regions, resulting in a slightly lower cosmic radiation flux at the equator. This geomagnetically trapped radiation consists of protons and electrons produced by primary cosmic radiation bombardment on the atmosphere. Because of the differences in mass and in electrical charge between protons and electrons, the trajectories of these particles diverge. As a result, two radiation belts form around the earth. One forms within 2.8 earth radii at the equator, and one forms outside of the same distance. Greater intensities and energies occur in the outer zone.<sup>12</sup> The inner radiation belt consists of protons, and the outer radiation belt consists of electrons. These belts of trapped radiation do not provide any appreciable exposures to the population of the earth.

Factors such as altitude and latitude contribute to variations to exposures from background radiation. Because of the magnetic properties of the earth, the charged particles originating as cosmic radiation are deflected away from the equator and are channeled to the poles. Primary cosmic radiation intensity remains fairly constant between 15°N and 15°S. Intensity increases rapidly to approximately 50°N and 50°S, after which it remains essentially constant again to the poles.<sup>13</sup>

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<sup>10</sup> Merrill Eisenbud and Thomas Gesell, Environmental Radioactivity: From Natural, Industrial, and Military Sources, 4<sup>th</sup> ed., (New York: Academic Press, 1997) 185.

<sup>11</sup> United Nations Scientific Committee on the Effects of Atomic Radiation, Sources and Effects of Ionizing Radiation, (New York: United Nations Publication, 1993) 36.

<sup>12</sup> *Ibid.*, 35.

<sup>13</sup> Eric J Hall, "Radiation and Life," April-1998, [<http://www.uic.com.au/ral.htm>].

Shielding effects from cosmic rays can be ignored because these particles are highly penetrating. The production rate at sea level of cosmic ray ions at middle latitudes and high latitudes falls within the range of approximately 1.9 ions/cm<sup>3</sup>/s to 2.6 ions/cm<sup>3</sup>/s, with an average of 2.1 ions/cm<sup>3</sup>/s.<sup>14</sup> Because low energy charged particles are deflected by the planetary magnetic field, a greater flux of protons reaches the poles than the equatorial regions. In addition, exposures to cosmic radiation will vary by an approximate factor of 2 over the range of elevation that encompasses most of the population of the world, from sea level to 2000 m.<sup>15</sup>

### **3.22. Cosmogenic Radionuclides**

Reactions between gases in the upper atmosphere and cosmic radiation produce cosmogenic radionuclides. These radionuclides form primarily through bombardment of the upper atmosphere by high energy heavy particles. Such cosmogenic radionuclides include tritium, <sup>14</sup>C, <sup>7</sup>Be, and <sup>22</sup>Na. Only tritium and <sup>14</sup>C really contribute to any significant exposures to the worldwide population. The exposures from these sources are relatively low and uniform over the surface of the planet.<sup>16</sup>

<sup>14</sup>C is present in carbon dioxide in the air, in the terrestrial biosphere, and in bicarbonates in the ocean. This radionuclide is produced in the atmosphere by the <sup>14</sup>N capture of neutrons. The neutron spectrum covers a wide energy range in the lower atmosphere, from thermal to 100 MeV.<sup>17</sup> The incident neutron flux over the planet is approximately 1 neutron/cm<sup>2</sup>/s. This neutron flux provides a natural, constant rate of production of <sup>14</sup>C, which results in a fairly constant concentration of <sup>14</sup>C in all biota of 6 pCi/g.<sup>18</sup> Equilibrium concentrations of <sup>14</sup>C in the atmosphere mainly are controlled through the exchange of carbon dioxide between the atmosphere and the oceans. The oceans provide a sink for removal of <sup>14</sup>C from the atmosphere.

An overwhelming majority of atmospheric nuclear weapons testing was conducted by the United States and the Soviet Union during the cold war. These tests peaked during 1955 to 1965. These tests also introduced large quantities of tritium into the atmosphere. Tritium behaves chemically as stable hydrogen gas right after creation. Initially, this radionuclide exists as HT, DT, or T<sub>2</sub>. Right after formation, however, most tritium oxidizes and forms tritiated water vapor. Spontaneous formation of tritium occurs from interactions of cosmic rays with gases in the upper atmosphere. In the stratosphere, tritium is spontaneously generated at a rate of 2500 atoms of <sup>3</sup>H/m<sup>2</sup>/s, and in the troposphere, tritium is generated at a rate of 5900 atoms of <sup>3</sup>H/m<sup>2</sup>/s.<sup>19</sup> These interactions result in an equilibrium concentration of tritium in the environment of 34 x 10<sup>6</sup> Ci.

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<sup>14</sup> UNSCEAR, 37.

<sup>15</sup> Committee on Evaluation of EPA Guidelines for Exposure to Naturally Occurring Radioactive Materials, *et. al.*, Evaluation of Guidelines for Exposures to Technologically Enhanced Naturally Occurring Radioactive Materials, (Washington, DC: National Academy Press, 1998) 2-1.

<sup>16</sup> B. G. Bennett, "Exposures to Natural Radiation Worldwide," High Levels of Natural Radiation 1996: Radiation Dose and Health Effects, (Amsterdam: Elsevier Science B. V., 1997) 18.

<sup>17</sup> UNSCEAR, 36.

<sup>18</sup> Eisenbud, 181.

<sup>19</sup> Rumi Nishimura, *et. al.*, "Evaluation of the Japanese Health Risks Induced by Global Fallout of

Time (y)	Energy Release (MeV)	Ion Pairs Produced
1.	$1. \times 10^6$	$32. \times 10^9$
50.	$55. \times 10^6$	$2. \times 10^{12}$

**TABLE 3.21: RELEASES OF IONIZING RADIATION AND ION PAIR PRODUCTION OF  $^{14}\text{C}$  IN BIOTA<sup>20</sup>**

Time (y)	Energy Release (MeV)	Ion Pairs Produced
1.	$718. \times 10^{21}$	$21. \times 10^{27}$
50.	$12. \times 10^{24}$	$363. \times 10^{27}$

**TABLE 3.22: RELEASES OF IONIZING RADIATION AND ION PAIR PRODUCTION OF  $^3\text{H}$  IN THE ENVIRONMENT<sup>21</sup>**

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*Tritium*, "Health Physics, vol. 75, no. 3, (September-1998): 259-68.

<sup>20</sup> Based on an initial activity of 6 pCi/g.

<sup>21</sup> Based on the equilibrium concentration of  $34 \times 10^6$  Ci.



### **3.30. Terrestrial Sources of Radiation**

Terrestrial sources of ionizing radiation originate from radionuclides that formed during the birth of the planet, as well as the secondary radionuclides derived from subsequent decay. These decay with long half lives. Two chains of terrestrial radionuclides contribute to a large majority of the background radiation present throughout the world: the uranium series ( $^{238}\text{U}$ ), and the thorium series ( $^{232}\text{Th}$ ). Exposures to these series of radionuclides vary widely.<sup>22</sup>

### **3.31. Variations**

Variations in background radiation can occur from such factors as soil moisture content and snow cover. Snow will act as a shield from radiation exposures at ground level. Therefore, lower background exposures will occur during the winter. Other seasonal changes also occur from the deposition of cosmogenic radionuclides on the ground. Deposition of radionuclides on the ground from the atmosphere is greater during the spring months when air in the stratosphere mixes with air in the troposphere. The radionuclides wash out of the air masses as a result of precipitation.<sup>23</sup> Conversely, high pressure systems provide extra shielding from cosmic radiation due to the presence of large air masses.

#### **3.31.1. Rock**

Terrestrial radionuclides are distributed throughout the crust of the earth. Outdoor exposures from sources of terrestrial radiation originate predominantly from approximately the top 1 ft layer of the soil.<sup>24</sup> Formations such as limestone and sandstone have the lowest radionuclide concentrations. Some shale formations, particularly those containing a significant fraction of organic matter, can possess higher concentrations of radionuclides. Overall, igneous and metamorphic rock comprise approximately 90% of the planetary crust. Sedimentary rocks, however, tend to accumulate at the top of the crust. Approximately 75% of the surface of the earth is covered by sedimentary rock.

The cooling and differentiation of molten magma results in the formation of silicate minerals. Igneous rocks are formed in the latter stages of this magmatic cooling and differentiation. These rocks contain mostly silicon and aluminum. When magma cools, minerals form that are rich in elements such as uranium, thorium, and potassium. The concentrations of these radionuclides are related to the amounts of silicates present during the formation of igneous

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<sup>22</sup> Bennett, 18.

<sup>23</sup> Phil Egidi, "Mobilization and Redeposition of NORM," 17-June-1998, [<http://www.normis.com/mob.htm>].

<sup>24</sup> K. G. Vohra, eds. *et. al.*, Natural Radiation Environment, (New York: John Wiley & Sons, 1982) 206.

rock. Acidic rock will contain the highest concentrations of radionuclides, while ultrabasic rock will contain the lowest concentrations of radionuclides.

Several types of processes will erode rock. If mechanical processes break down igneous rock, then a separation will occur to divide major and minor silicates. The minor silicates are rich in terrestrial radionuclides. Such minor silicates include zircon and monazite. These minerals are stable and resistant to chemical degradation. If chemical or biological processes break down igneous rock, then the minor silicates usually release the radionuclides into layers of clay minerals. When the igneous rocks completely erode, the radionuclides remain, trapped in the layers of clay.

A large majority of land area in the continental United States consists of sedimentary rock. Sedimentation processes naturally sort radionuclide inventories, subsequently forming rock types of differing radionuclide concentrations. Shale and phosphate rock usually have the highest concentrations of radionuclides. In addition, uranium and thorium can become mobile in sedimentary rock through the migration of water or of oil.

Shale is comprised of at least 35% clay. A significant fraction of shale contains potassium. Shale also can adsorb uranium and thorium. These radionuclides typically are bound to organic matter in minerals in the rock. The uranium and thorium also can be found as precipitates in the material that binds the rock. Quartz is the major mineral that comprises sandstone. Some sandstone contains feldspar in fractions as high as 25%. Feldspar is usually enriched with potassium. Concentrations of uranium and thorium are low in sandstone. Deposits of these radionuclides have been found at the boundaries of different layers of sandstone.

Carbonate rock is found as limestone or as dolomite, which is formed as a result of the precipitation of materials such as bones and shells from water. Generally, carbonate itself is almost free of radionuclides. The spaces between carbonate grains, however, can contain some radionuclides. Potassium is very soluble in sea water, and it can remain in these interstitial spaces as the sea water is removed. Thorium also can be present, as it is found in depleted concentrations in sea water. Uranium can be found in limestone as a result of the decay of organic matter deposited where the limestone is located.

### 3.31.2. Soil

Sources of ionizing radiation found in soil results from the rock from which the soil is derived. Levels of ionizing radiation will vary as a result of diminishment by leaching of water, dilution by increased porosity, and by augmentation from water and organic matter by sorption and precipitation of radionuclides.<sup>25</sup>

Radionuclide	Mean Concentration (Bq/kg)	Range (Bq/kg)
<sup>40</sup> K	370	100 – 700
<sup>232</sup> Th series	35	4 – 130
<sup>238</sup> U series	35	4 – 140
<sup>226</sup> Ra subseries	40	8 – 160

TABLE 3.33: CONCENTRATIONS OF NATURAL RADIONUCLIDES IN SOIL<sup>26</sup>

Radionuclide	Energy Release (MeV)		Ion Pairs Produced	
	Mean	Range	Mean	Range
<sup>40</sup> K	774. x 10 <sup>9</sup>	209. x 10 <sup>9</sup> – 1. x 10 <sup>12</sup>	23. x 10 <sup>15</sup>	6. x 10 <sup>15</sup> – 43. x 10 <sup>15</sup>
<sup>232</sup> Th series	238. x 10 <sup>9</sup>	27. x 10 <sup>9</sup> – 883. X 10 <sup>9</sup>	7. x 10 <sup>15</sup>	799. x 10 <sup>12</sup> – 26. x 10 <sup>15</sup>
<sup>238</sup> U series	1. x 10 <sup>12</sup>	116. x 10 <sup>9</sup> – 4. x 10 <sup>12</sup>	30. x 10 <sup>15</sup>	3. x 10 <sup>15</sup> – 120. x 10 <sup>15</sup>
<sup>226</sup> Ra series	304. x 10 <sup>9</sup>	61. x 10 <sup>9</sup> – 1. x 10 <sup>12</sup>	9. x 10 <sup>15</sup>	2. x 10 <sup>15</sup> – 36. x 10 <sup>15</sup>

TABLE 3.33.1: RELEASES OF IONIZING RADIATION FROM NATURAL RADIONUCLIDES IN SOIL AT 50 YEARS

<sup>25</sup> D. J. Beninson, *et. al.*, “*Dosimetric Implications of the Exposure to the Natural Sources of Irradiation*,” Simpósio Internacional Sobre Áreas De Elevada Radioatividade Natural, (Rio de Janeiro, Brazil: Academia Brasileira de Ciências, 1977) 80.

<sup>26</sup> UNSCEAR, 65.

Several factors contribute to the composition of the soil, and subsequently, to the concentrations of radionuclides in the soil. Soil can be formed from the erosion of subsurface bedrock. Soil also can be formed as a result of the transportation and the weathering of different rock types. Transport mechanisms involve natural phenomena such as earthquakes, volcano eruptions, major floods, and glacial movements. Such glacial deposits are common in areas such as the Great Lakes, New England, and Alaska. Outwash erosion products from mountains can form soil surfaces that are more radioactive than underlying bedrock.<sup>27</sup>

### **3.31.3. Mobilization**

Both man made and natural processes factor into the mobilization of radionuclides. Some radionuclides will dissolve into groundwater. Air flow serves as a transport mechanism for fine radionuclide particulates. Other physical and chemical processes involve combustion and volatilization. Uranium extraction will maximize its solubility. Extraction of phosphate from ore will result in high concentrations of uranium remaining in a filter cake. Combustion of coal leaves behind high uranium concentrations in ash.

Changes in the chemistry of groundwater can lead to mobilization and deposition. As groundwater travels through different layers of rock and of soil, changes can result in pH, oxidation state, temperature, or chemical equilibrium of the groundwater. Such changes will affect the solubility of minerals in the groundwater. These conditions can lead to the precipitation of minerals enriched with radionuclides. In addition, the passage of groundwater through materials such as clay sometimes will result in the adsorption of radionuclides.

### **3.32. Uranium**

Three uranium isotopes occur in nature:  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ . The isotope of  $^{238}\text{U}$  occurs with a natural abundance of 99.28%,  $^{235}\text{U}$  occurs with a natural abundance of .71%, and  $^{234}\text{U}$  occurs with a natural abundance of .0058%. Uranium is prevalent to some degree in all common types of rock and soil. Common rock types contain concentrations of uranium in the range of .5 ppm to 4.7 ppm. These concentrations, however, do not only just refer to  $^{238}\text{U}$  itself, but also to the daughter products inherently contained in the uranium decay chain. Each radionuclide in this decay chain will emit several different types of radioactive particles and photons. Over a sufficiently long time, the decay chain essentially behaves as a single, large source of ionizing radiation. Considerable energy releases occur as a result of the decay of this natural series.

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<sup>27</sup> Phil Egidi, "Geology of NORM," 17-June-1998, [<http://www.normis.com/geo.htm>].

Material	Uranium-238			
	ppm	Bq/kg	Energy Release (MeV)	Ion Pairs Produced
<b>Igneous Rock</b>				
Basalt (crustal avg)	.5 – 1	7 – 10	$203. \times 10^9 - 291. \times 10^9$	$6. \times 10^{15} - 9. \times 10^{15}$
Mafic	.5, .9	7, 10	$203. \times 10^9, 291. \times 10^9$	$6. \times 10^{15}, 9. \times 10^{15}$
Salic	3.9, 4.7	50, 60	$1. \times 10^{12}, 2 \times 10^{12}$	$43. \times 10^{15}, 51. \times 10^{15}$
Granite (crustal avg)	3	40	$1. \times 10^{12}$	$34. \times 10^{15}$
<b>Sedimentary Rock</b>				
Shale	3.7	40	$1. \times 10^{12}$	$34. \times 10^{15}$
Clean Quartz	< 1	< 10	$< 291. \times 10^9$	$< 9. \times 10^{15}$
Dirty Quartz	2 – 3 ?	40 ?	$1. \times 10^{12} ?$	$34. \times 10^{15} ?$
Arkose	1 – 2 ?	10 – 25 ?	$291. \times 10^9 - 726. \times 10^9 ?$	$9. \times 10^{15} - 21. \times 10^{15} ?$
Beach Sand	3	40	$1. \times 10^{12}$	$34. \times 10^{15}$
Carbonate Rock	2	25	$726. \times 10^9$	$21. \times 10^{15}$
Continental Crust (avg)	2.8	36	$1. \times 10^{12}$	$31. \times 10^{15}$
Soil (avg)	1.8	22	$639. \times 10^9$	$19. \times 10^{15}$

TABLE 3.34: RELEASES OF IONIZING ENERGY IN TYPICAL ROCK AT 50 YEARS<sup>28</sup>

The concentrations of uranium in public water supplies varies greatly throughout the country. The two main valence states of uranium that are stable in geological environments are the uranous ( $U^{4+}$ ) and uranyl ( $U^{6+}$ ) states. Uranous is essentially insoluble. Uranium transport generally occurs in oxidizing surface water and groundwater as the uranyl ion ( $UO_2^{2+}$ ), uranyl fluoride, uranyl carbonate complexes, or uranyl phosphate.<sup>29</sup> The presence of uranyl phosphate results in prevalent concentrations of uranium in phosphate fertilizers. In addition, this radionuclide is present in food and human tissues. The annual intake of uranium from all dietary sources averages approximately 320 pCi (13 Bq).<sup>30</sup> The intake of 320 pCi of uranium will result in the committed energy release of 11000 MeV of ionizing radiation over a 50 year period.

<sup>28</sup> National Council on Radiation Protection and Measurements, "Exposure of the Population in the United States and Canada From Natural Background Radiation," NCRP Report No. 94, (30-December-1987): National Council on Radiation Protection and Measurements, Bethesda, MD, 61.

<sup>29</sup> Committee on Evaluation of EPA Guidelines, 2-3.

<sup>30</sup> Eisenbud, 141.

### 3.32.1 Oklo

The open pit uranium mine located in the West African republic of Gabon has been mined by the French nuclear industry for decades. In 1972, a shipment of uranium was found to have depleted concentrations of  $^{235}\text{U}$ . After further investigation, scientists discovered that the Oklo mine contained the remains of a naturally occurring nuclear reactor.

Currently,  $^{235}\text{U}$  only exists in nature with an abundance of .72%. Nuclear power reactors in this country enrich nuclear fuel to approximately 3%  $^{235}\text{U}$ . Based on the half life of  $^{235}\text{U}$ , which is  $704 \times 10^6$  y, the abundance of this radionuclide in nature was approximately 3% around 2 billion years ago, during the Proterozoic Era. Uranium most likely originated from igneous deposits, located near the pit. Mobilization through surface water and groundwater then occurred when sufficient concentrations of dissolved oxygen became available in the system. In the presence of oxygen, the uranous ion ( $\text{U}^{4+}$ ) will bond to dissolved oxygen in water, thus forming the mobile uranyl ion  $\text{UO}_2^{2+}$ . The infiltration of uranium into groundwater and surface water allowed for the transport of this radionuclide to the Oklo pit 2 billion years ago.

Subsequent reactions with organic matter concentrated the dissolved uranium oxide into the pure form. When a critical mass was achieved, the chain reaction commenced. Water infiltration through porous rocks in the area not only carried the nuclear fuel to this natural core, but provided moderation and reflection as well. The reactor is thought to have operated for a period in excess of  $10^5$  years, with a thermal neutron flux in the range of  $10^9$  n/cm<sup>2</sup>/s to  $10^{21}$  n/cm<sup>2</sup>/s.<sup>31</sup> An estimated 15000 MW-years of energy would have been released through the consumption of 6000 kg of  $^{235}\text{U}$ .<sup>32</sup> This scale of fuel consumption would release ionizing radiation to the environment at a total of approximately  $3. \times 10^{30}$  MeV, based on the total life time of the reactor system.

The natural reactor could not have operated continuously for such a long period of time. The ionizing radiation that was released as a result of this natural fission most likely dissipated the water present at the site, thus reducing moderation, causing the chain reaction to cease. A restart of the reactor would not have occurred until the reactor pit was saturated again, and when fission product poisons decayed fully.

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<sup>31</sup> Andrew Karam, "The Natural Nuclear Reactor at Oklo: A Comparison with Modern Nuclear Reactors," 17-November-1998, [<http://www.physics.isu.edu/radinf/oklo.htm>].

<sup>32</sup> *Ibid.*

### 3.33. Radium

Radium is another important radioisotope contributing to natural radiation exposures to humans. Ingestion is the primary route for transfer of radium to the human body.<sup>33</sup> <sup>226</sup>Ra is a long lived, alpha particle emitting radionuclide. Concentrations of this radionuclide can be found in all common rock and soil. Average concentrations of radium in limestone and in igneous rock are .42 pCi/g, and 1.3 pCi/g, respectively.<sup>34</sup>

Radium exists in most water supplies. The current EPA limit of radium concentration in water (5 pCi/L), has been exceeded in some smaller communities that obtain potable water from wells.<sup>35</sup> Approximately 75% of the groundwater supplies in the United States that exceed the EPA National Primary Interim Drinking Water Standard for radium of 5 pCi/L were found to located in two areas: the piedmont and coastal plain areas of the middle atlantic states and the north central states of Minnesota, Iowa, Illinois, Missouri, and Wisconsin.

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<sup>33</sup> Bennett, 18.

<sup>34</sup> Eisenbud, 143.

<sup>35</sup> *Ibid.*, 144.

Rock Type	Mean Concentration (Bq/kg)	Range (Bq/kg)
Acid Intrusive	78	1 – 370
Basic Extrusive	11	.4 – 41
Chemical Sedimentary	45	.4 – 340
Detrital Sedimentary	60	1 – 990
Metamorphosed Igneous	50	1 – 1800
Metamorphosed Sedimentary	37	1 – 660

TABLE 3.35: CONCENTRATIONS OF  $^{226}\text{Ra}$  IN VARIOUS TYPES OF ROCK<sup>36</sup>

Rock Type	Energy Release (MeV)		Ion Pairs Produced	
	Mean	Range	Mean	Range
Acid Intrusive	$593. \times 10^9$	$8. \times 10^9 - 3. \times 10^{12}$	$17. \times 10^{15}$	$224. \times 10^{12} - 83. \times 10^{15}$
Basic Extrusive	$84. \times 10^9$	$3. \times 10^9 - 312. \times 10^9$	$2. \times 10^{15}$	$89. \times 10^{12} - 9. \times 10^{15}$
Chemical Sedimentary	$342. \times 10^9$	$3. \times 10^9 - 3. \times 10^{12}$	$10. \times 10^{15}$	$89. \times 10^{12} - 76. \times 10^{15}$
Detrital Sedimentary	$456. \times 10^9$	$3. \times 10^9 - 8. \times 10^{12}$	$13. \times 10^{15}$	$224. \times 10^{12} - 221. \times 10^{15}$
Metamorphosed Igneous	$380. \times 10^9$	$3. \times 10^9 - 14. \times 10^{12}$	$11. \times 10^{15}$	$224. \times 10^{12} - 402. \times 10^{15}$
Metamorphosed Sedimentary	$281. \times 10^9$	$3. \times 10^9 - 5. \times 10^{12}$	$8. \times 10^{15}$	$224. \times 10^{12} - 148. \times 10^{15}$

TABLE 3.35.1: RELEASES OF IONIZING RADIATION IN VARIOUS TYPES OF ROCK AT 50 YEARS

<sup>36</sup> UNSCEAR, 69.



### 3.33.1. Bone

Radium and radium progeny contribute to a major fraction of internal exposures to humans. Radium is more chemically active than most other radionuclides. Since radium is chemically similar to calcium, this radionuclide can be absorbed from the soil by plants and, subsequently, ingested by humans. Typical  $^{226}\text{Ra}$  levels in most components of the diet range from .1 pCi/kg to 1 pCi/kg.<sup>37</sup> When ingested, radium replaces calcium in bone with a biological systemic uptake of approximately 20%. A large majority of the total body content (80% - 85%) of radium is located in the skeleton. Average concentrations of radium in an adult skeleton containing 1 kg of calcium range between 8 pCi (.3 Bq) to 100 pCi (3.7 Bq), with a population weighted skeletal content of 23 pCi (.85 Bq).<sup>38</sup>

Radioisotope	Activity	Energy Release (MeV)	Ion Pairs Produced
Radium	8 pCi – 100 pCi	$2. \times 10^9 - 28. \times 10^9$	$66. \times 10^{12} - 828. \times 10^{12}$
	.1 pCi/kg – 1 pCi/kg	$28. \times 10^6 - 282. \times 10^6$	$828. \times 10^9 - 8. \times 10^{12}$
	23 pCi	$6. \times 10^9$	$190. \times 10^{12}$

TABLE 3.36: RELEASES IONIZING RADIATION IN THE BODY AT 50 YEARS

<sup>37</sup> D. J. Beninson, *et. al.*, "Dosimetric Implications of the Exposure to the Natural Sources of Irradiation," Simpósio Internacional Sobre Áreas De Elevada Radioatividade Natural, (Rio de Janeiro, Brazil: Academia Brasileira de Ciencias, 1977) 93.

<sup>38</sup> Eisenbud, 149.

### 3.34. Thorium

Like uranium and radium, thorium is found in all types of rock and soil. Concentrations of thorium in common rock types fall between 1.6 ppm and 20 ppm. The concentrations of thorium in rock and in soil generally exceed that of uranium. Since the specific activity of thorium is slightly less than the specific activity of uranium, the exposures to ionizing radiation from both of these radionuclides are approximately equal.

Thorium is essentially insoluble. Therefore, concentrations of this radionuclide in biological material is almost negligible, although small amounts of thorium are evident in some vegetables. This radionuclide also is not mobile in the environment. The highest concentrations of thorium in the body have been found predominantly in the pulmonary lymph nodes and lungs. The presence of high concentrations in this area of the body indicates that infiltration occurs mainly as a result of inhalation of soil and dust particles.

Material	Thorium-232			
	ppm	Bq/kg	Energy Release (MeV)	Ion Pairs Produced
<b>Igneous Rock</b>				
Basalt (crustal avg)	3 – 4	10 – 15	$68. \times 10^9 - 102. \times 10^9$	$2. \times 10^{15} - 3. \times 10^{15}$
Mafic	1.6, 2.7	7, 10	$48. \times 10^9, 102. \times 10^9$	$1. \times 10^{15}, 3. \times 10^{15}$
Salic	16, 20	60, 80	$408. \times 10^9, 543. \times 10^9$	$12. \times 10^{15}, 16. \times 10^{15}$
Granite (crustal avg)	17	70	$476. \times 10^9$	$14. \times 10^{15}$
<b>Sedimentary Rock</b>				
Shale	12	50	$340. \times 10^9$	$10. \times 10^{15}$
Clean Quartz	< 2	< 8	$< 54. \times 10^9$	$< 2. \times 10^{15}$
Dirty Quartz	3 – 6 ?	10 – 25 ?	$102. \times 10^9 - 170. \times 10^9 ?$	$3. \times 10^{15} - 5. \times 10^{15} ?$
Arkose	2 ?	< 8	$< 54. \times 10^9$	$< 2. \times 10^{15}$
Beach Sand	6	25	$170. \times 10^9$	$5. \times 10^{15}$
Carbonate Rock	2	8	$54. \times 10^9$	$2. \times 10^{15}$
Continental Crust (avg)	10.7	44	$299. \times 10^9$	$9. \times 10^{15}$
Soil (avg)	9	37	$251. \times 10^9$	$7. \times 10^{15}$

TABLE 3.37: RELEASES OF IONIZING RADIATION TYPICAL ROCK AT 50 YEARS<sup>39</sup>

<sup>39</sup> National Council on Radiation Protection and Measurements, 61.

### 3.35. Radon

Radon exposures represent the largest fraction of total background radiation. Radon is a noble gas. Therefore, radon gas does not chemically react with other elements or materials in the environment. Several isotopes of radon exist in nature.  $^{219}\text{Rn}$  exists as a result of the decay of  $^{235}\text{U}$ . This isotope of uranium, however, occurs in nature with an abundance of less than 1%. In addition,  $^{219}\text{Rn}$  decays with a half life of approximately 4 seconds. The decay of  $^{232}\text{Th}$  leads to the formation of  $^{220}\text{Rn}$ . The natural abundance of  $^{232}\text{Th}$  is approximately 99%, nearly equal to the natural abundance of  $^{238}\text{U}$ , the parent radionuclide of  $^{222}\text{Rn}$ . As a result, the production of  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$  are about equal. The half life of  $^{220}\text{Rn}$ , however, is 55 seconds. The half life of  $^{222}\text{Rn}$  is approximately 4 days. Therefore,  $^{220}\text{Rn}$  and its daughter products are usually neglected when dealing with the issue of radon exposures.<sup>40</sup> The most significant mechanism of exposure to radon is the inhalation of the short lived decay products of the parent radionuclide.

#### 3.35.1. Outdoor Radon

The main source of radon concentrations in outdoor air originates from soil emanation. Radium content in soil, as well as soil properties, are determining factors that affect radon concentrations, but moisture content and meteorological conditions are also of influence.<sup>41</sup> Granite rock has the highest concentrations of radium, and basalt rock has the lowest concentrations of radium. In order to migrate, radon first must emanate from mineral substances into the pore spaces of the soil matrix. This process is brought about by the alpha particle recoil of radium atoms on formation of radon, which have a range of 20 nm to 70 nm, and by molecular diffusion.<sup>42</sup> When the radium atom ejects an alpha particle, the newly formed radon atom recoils in the opposite direction. Alpha particle recoil is the dominant factor governing the release of radon from mineral grains. Therefore, the location of the radium atom in the mineral grain, and the direction of recoil of the radon atom, will determine whether the newly formed radon atom will enter the pore spaces of the soil matrix. Approximately 10% to 50% of radon produced will migrate from the mineral grains to the soil matrix. Most soil in the United States contains between .33 pCi and 1 pCi of radium per gram of mineral matter, thus resulting in 200 pCi to 2000 pCi of radon per liter of soil gas.<sup>43</sup>

Since radon is a gas, this radionuclide possesses a far greater mobility than uranium or radium. The rate of radon mobility is dependent upon soil moisture content, soil porosity, and soil permeability. Therefore, radon can migrate easily through materials such as coarse sand or

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<sup>40</sup> David Bodansky, *et. al.*, eds., Indoor Radon and Its Hazards, (Seattle: University of Washington Press, 1987) 5.

<sup>41</sup> Bennett, 19.

<sup>42</sup> UNSCEAR, 45.

<sup>43</sup> United States Geological Survey, "The Geology of Radon," 13-October-1995, [<http://sedwww.cr.usgs.gov:8080/radon/georadon/3.html>].

gravel. In addition, the presence of water results in larger quantities of radon available for movement because more recoil atoms will terminate in the pore spaces rather than in the soil matrix. Radon will migrate slower, however, in water than in soil gas. Therefore, houses in areas with dry, highly permeable soil and rock will have a higher indoor radon concentration.

The movement of radon through porous materials is governed by concentration differences and by pressure gradients. The mechanisms of radon movement are diffusion and convection. The diffusion process dominates over convection as the mechanism by which radon enters the atmosphere from the surface of the earth, with an estimated volumetric production rate of  $.027 \text{ Bq/m}^3/\text{s}$ , and an estimated flux of approximately  $.026 \text{ Bq/m}^2/\text{s}$ .<sup>44</sup>

Radioisotope	Activity	Energy Release (MeV)	Ion Pairs Produced
Radon	200 – 2000 pCi/L	$1. \times 10^9 - 11. \times 10^9$	$32. \times 10^{12} - 317. \times 10^{12}$

**TABLE 3.38: RELEASES FROM RADON IN OUTDOOR AIR AT 50 YEARS**

Concentrations of radon and radon daughter products in air are extremely variable. Temporal changes contribute greatly to the high variability in background exposures. Concentrations of radon in the air fluctuate on a daily basis. Diurnal variations in radon concentrations in air at ground level occur with a minimum at noon and a maximum two times higher at night.<sup>45</sup> In the morning, air is colder and heavier. Therefore, concentrations remain high. During the day, increasing temperatures cause the air to heat and rise, thus allowing radon to migrate to the upper atmosphere, reducing ambient concentrations. Atmospheric concentrations of radon are much less than soil concentrations, primarily because of dispersion. Data from the United States and several other countries indicate that the average concentrations of radon in outdoor air are approximately  $.1 \text{ pCi/L}$  ( $4 \text{ Bq/m}^3$ ) to  $.5 \text{ pCi/L}$  ( $19 \text{ Bq/m}^3$ ).<sup>46</sup> Concentrations vary as a result of specific meteorological conditions in each locality. Conditions such as barometric pressure can affect the rate of gas exchange at the interface of the soil and the atmosphere. Precipitation tends to reduce the concentration of radon in the air because higher quantities of radon will dissolve in rainwater and will infiltrate the soil. Conversely, periods of temperature inversion will increase atmospheric content of radon because lesser volumes of air will be present in which dilution and dispersion can take place.

<sup>44</sup> UNSCEAR, 46.

<sup>45</sup> Bennett, 19.

<sup>46</sup> Committee on Evaluation of EPA Guidelines for Exposure to Naturally Occurring Radioactive Materials, 2-8.

Elevated radon concentrations in potable water supplies provides another mechanism for human exposures. Large, open water sources such as reservoirs have a low radon content because radon will escape into the atmosphere. Municipal water supplies also do not have a high radon content because these systems usually aerate the water, thus allowing radon to escape. Large, public groundwater supplies generally are drawn from sedimentary rock sources. These formations have low radium content; consequently, such groundwater supplies are low in radon content. Small, public water supplies and private wells operate with closed systems. Therefore, radon content can be quite high in these supplies, as the radionuclide will not diffuse into the atmosphere. Studies of small community supplies have found levels that range between  $10^3$  kBq/m<sup>3</sup> -  $10^4$  kBq/m<sup>3</sup> ( $10^4$  pCi/L -  $10^5$  pCi/L).<sup>47</sup> EPA estimates show that 17 million Americans are served by systems that provide water containing concentrations of radon in excess of 300 pCi/L.<sup>48</sup>

Radioisotope	Activity	Energy Release (MeV)	Ion Pairs Produced
<sup>222</sup> Rn	.1 – .5 pCi/L (in outdoor air)	$539. \times 10^3 - 3. \times 10^6$	$16. \times 10^9 - 79. \times 10^9$
	$10^4$ pCi/L – $10^5$ pCi/L	$54. \times 10^9 - 539. \times 10^9$	$2. \times 10^{15} - 16. \times 10^{15}$
	300 pCi/L	$2. \times 10^9$	$48. \times 10^{12}$

**TABLE 3.39: RELEASES OF IONIZING RADIATION INTO POTABLE WATER SUPPLIES AT 50 YEARS**

<sup>47</sup> Eisenbud, 157.

<sup>48</sup> *Ibid.*, 157.

### 3.35.2. Indoor Radon

The largest and most variable component of exposures to natural sources of ionizing radiation is due to radon and its decay products in indoor air.<sup>49</sup> Radon enters indoor air from soil or from rock surrounding a dwelling, as well as from building materials and water supplies. The ventilation rate is an important factor governing the concentration of radon indoors. Radon infiltration to the indoors occurs mainly in residential areas. Such dwellings are usually small, containing only a maximum of two stories. Because the radon originates mostly from the soil, the radionuclide has the greatest effect on these smaller structures. Radon can infiltrate a house through diffusion. Infiltration of radon into houses depends upon the amount of the radionuclide present in pores in the soil matrix and in rock fractures. Transport of the gas is governed by the size of the pore spaces within the soil matrix as well as the permeability of the soil. The radionuclide can diffuse from the soil through any breaches present in the foundation. A discontinuity of only 1% in a floor can allow a 20% increase in the rate of diffusion of radon into a house.<sup>50</sup>

The rate of infiltration also depends upon the materials used for the floors and for the walls. The rate of radon infiltration by diffusion from bare earth is estimated to be approximately 37 Bq/m<sup>3</sup>/s, but, with the addition of a concrete floor, that rate is reduced to 2.6 Bq/m<sup>3</sup>/s.<sup>51</sup> Board floors really do not impede the infiltration of radon indoors. Some rock types that usually have large fracture spaces are granite, marine shale, and recrystallized limestone. Soils consisting of coarse grain sizes will have large pore spaces and high permeabilities.

Infiltration rates will vary greatly with the condition of the floor and the permeability of the soil. Entry rates vary from zero for an impermeable floor element, to 10 Bq/m<sup>3</sup>/s, for a cracked floor on soil with a low permeability, and to 274 Bq/m<sup>3</sup>/s, for a bare floor with a soil of high permeability.<sup>52</sup> Factors such as structural details of the building, as well as the characteristics of soil and of rock contribute to the high variability of radon concentrations and radon infiltration rates. In addition, buildings with basements will show an increase in infiltration rates as a result of the greater surface area in contact with the soil and the rock.

Convection also affects the rate of radon infiltration to the indoor environment. The wind blowing on a building and the heating system inside the building provide the two dominant mechanisms for convection. Changes in barometric pressure and in mechanical ventilation also provide other minor factors contributing to the overall convection of the indoor environment. Convection will dominate over diffusion as the mechanism for radon infiltration into the indoor environment under normal and average circumstances.

Several factors contribute to the variability of indoor radon concentrations. Meteorological factors can affect concentrations indoors in the same manner that such factors

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<sup>49</sup> Bennett, 16.

<sup>50</sup> UNSCEAR, 47.

<sup>51</sup> *Ibid.*, 47.

<sup>52</sup> *Ibid.*, 47.

influence outdoor concentrations. Higher concentrations in the soil present more quantities of radon available for transport indoors. Areas with high concentrations in groundwater can lead to high concentrations indoors as well. The choice of building materials also can hinder or can facilitate radon infiltration.

Radioisotope	Activity	Energy Release (MeV)	Ion Pairs Produced
$^{222}\text{Rn}$	10 – 274 Bq/m <sup>3</sup>	$1.4 \times 10^9 - 40. \times 10^9$	$43. \times 10^{12} - 1. \times 10^{15}$
	37 Bq/m <sup>3</sup>	$5. \times 10^9$	$158. \times 10^{12}$
	2.6 Bq/m <sup>3</sup>	$378. \times 10^6$	$11. \times 10^{12}$

TABLE 3.310: RELEASES OF IONIZING RADIATION TO INDOOR AIR AT 50 YEARS

### 3.35.3. Lead and Polonium

$^{210}\text{Pb}$  is the longest lived daughter in the radon decay chain. This radionuclide decays with a half life of approximately 22 years.  $^{210}\text{Pb}$  is produced rapidly in the atmosphere. This radionuclide usually will precipitate to the surface of the earth, in the form of rain or of snow. Only minute quantities of  $^{210}\text{Pb}$  will decay in the atmosphere, as a result of the long half life. Rainwater concentrations of  $^{210}\text{Pb}$  fall between 1 pCi/L (37 Bq/m<sup>3</sup>) and 10 pCi/L (370 Bq/m<sup>3</sup>), with a mean of approximately 2 pCi/L (74 Bq/m<sup>3</sup>).<sup>53</sup> Concentrations of  $^{210}\text{Pb}$  in a standard diet range from .70 pCi/kg (.026 Bq/kg) to 1.0 pCi/kg (.037 Bq/kg), with a worldwide average daily intake of 3 pCi (.11 Bq).<sup>54</sup>

$^{210}\text{Po}$  forms as a result of  $^{210}\text{Pb}$  decay. Very little of this radionuclide is produced in the atmosphere. At the ground level, both of these radionuclides usually attach to dust particles. Therefore, atmospheric transport provides the dominant mechanism for which  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  distribute at ground level. Intakes of lead and polonium can originate from atmospheric deposition of these radionuclides on broad leafed plants, such as tobacco.

When  $^{210}\text{Pb}$  is absorbed into the body, ingrowth of  $^{210}\text{Po}$  can occur because  $^{210}\text{Pb}$  will be deposited in the skeleton, from which it is removed slowly, with a biological half life of approximately  $10^4$  days.<sup>55</sup> The skeleton contains approximately 70% of the body burden of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . Some  $^{210}\text{Po}$  is absorbed by the blood and deposited in other tissues or excreted by the body. Exposures resulting from ingestion of these radionuclides dominates over inhalation.

<sup>53</sup> Eisenbud, 168.

<sup>54</sup> *Ibid.*, 168.

<sup>55</sup> *Ibid.*, 168.

Radionuclide	Activity	Energy Release (MeV)	Ion Pairs Produced
$^{210}\text{Pb}$	1 – 10 pCi/L	$3. \times 10^6 - 34. \times 10^6$	$100. \times 10^9 - 996. \times 10^9$
	.7 – 1 pCi/kg	$2. \times 10^6 - 3. \times 10^6$	$70. \times 10^9 - 100. \times 10^9$
	2 pCi/L	$7. \times 10^6$	$199. \times 10^9$
	3 pCi	$10. \times 10^6$	$299. \times 10^9$

TABLE 3.311: RELEASES OF IONIZING RADIATION FROM  $^{210}\text{Pb}$  IN RAINWATER AND IN THE STANDARD DIET AT 50 YEARS

### 3.35.4. Smoking

$^{210}\text{Pb}$  is deposited on the leaves of tobacco plants via atmospheric transport. Minute quantities also are deposited into the plant via uptake from the soil. The  $^{210}\text{Po}$  content of tobacco follows from ingrowth in  $^{210}\text{Pb}$ . Exposures to  $^{210}\text{Po}$  is highest for smokers, where inhalation of cigarette smoke will result in the deposition of these radionuclides on the tracheobronchial tree. Smoking ten cigarettes per day will increase the intake of  $^{210}\text{Po}$  by an approximate factor of two. These radionuclides can irradiate basal cells of the bronchial epithelium. Smoking one pack of cigarettes per day results in an intake of .3 pCi to .8 pCi of  $^{210}\text{Pb}$  and .4 pCi to 1.4 pCi of  $^{210}\text{Po}$ .<sup>56</sup> Exposures to the total skeleton are elevated by 30%, and exposures to the bone surfaced are increased by 8% for smokers.<sup>57</sup>

Radionuclide	Activity	Energy Release (MeV)	Ion Pairs Produced
$^{210}\text{Pb}$	.3 pCi – .8 pCi	$2. \times 10^6 - 4. \times 10^6$	$60. \times 10^9 - 118. \times 10^9$
$^{210}\text{Po}$	.4 pCi – 1.4 pCi	$1. \times 10^6 - 5. \times 10^6$	$40. \times 10^9 - 139. \times 10^9$
<b>Total</b>		<b><math>3. \times 10^6 - 9. \times 10^6</math></b>	<b><math>100. \times 10^9 - 257. \times 10^9</math></b>

TABLE 3.312: RELEASES OF IONIZING RADIATION IN THE BODY FROM  $^{210}\text{Pb}$  AND  $^{210}\text{Po}$  AT 50 YEARS<sup>58</sup>

<sup>56</sup> Environmental Protection Agency, "Radiological Quality of the Environment in the United States, 1977," EPA-520/1-77-009, (September-1977): Environmental Protection Agency, Washington, DC, 34.

<sup>57</sup> *Ibid.*, 170.

<sup>58</sup> These intakes are based upon smoking a pack of cigarettes per day.



### 3.36. Potassium

$^{40}\text{K}$  is present in nature with an abundance of approximately .0118% in natural potassium. Because  $^{40}\text{K}$  decays with a highly energetic negatron of 1.31 MeV, this radionuclide comprises the largest radioactive component in human tissues and in most foods. Most of the potassium in the human body is found in the muscle. At the age of thirty years, the body content of  $^{40}\text{K}$  is approximately .18%.<sup>59</sup> Therefore, a person weighing 150 lb will have approximately 130 g of potassium in the body. With an abundance of  $1.18 \times 10^{-4}$ , imparting a specific activity of 800 pCi/g, the activity of  $^{40}\text{K}$  in the body will be approximately .1  $\mu\text{Ci}$ . This activity leads to a 50 year committed exposure to ionizing radiation of  $8. \times 10^{12}$  MeV from  $^{40}\text{K}$  in the body. Potassium content in the body is under homeostatic control. Therefore, the concentration of  $^{40}\text{K}$  in the body does not fluctuate as a result of changes in environmental variables.

Material	Potassium-40			
	% total K	Bq/kg	Energy Release (MeV)	Ion Pairs Produced
<b>Igneous Rock</b>				
Basalt (crustal avg)	.8	300	$627. \times 10^9$	$18. \times 10^{15}$
Mafic	.3 – 1.1	70 – 400	$146. \times 10^9 - 836. \times 10^9$	$4. \times 10^{15} - 25. \times 10^{15}$
Salic	4.5	1100 – 1500	$2. \times 10^{12} - 3. \times 10^{12}$	$68. \times 10^{15} - 92. \times 10^{15}$
Granite (crustal avg)	> 4	> 1000	$2. \times 10^{12}$	$61. \times 10^{15}$
<b>Sedimentary Rock</b>				
Shale	2.7	800	$2. \times 10^{12}$	$49. \times 10^{15}$
Clean Quartz	< 1	< 300	$627. \times 10^9$	$18. \times 10^{15}$
Dirty Quartz	2 ?	400 ?	$836. \times 10^9$	$25. \times 10^{15}$
Arkose	2 – 3	600 – 900	$1. \times 10^{12}$	$55. \times 10^{15}$
Beach Sand	< 1	< 300	$627. \times 10^9$	$18. \times 10^{15}$
Carbonate Rock	.3	70	$146. \times 10^9$	$4. \times 10^{15}$
Continental Crust (avg)	2.8	850	$2. \times 10^{12}$	$52. \times 10^{15}$
Soil (avg)	1.5	400	$836. \times 10^9$	$25. \times 10^{15}$

TABLE 3.313: RELEASES OF IONIZING RADIATION OF TYPICAL ROCK AT 50 YEARS<sup>60</sup>

<sup>59</sup> UNSCEAR, 43.

<sup>60</sup> National Council on Radiation Protection and Measurements, 61.

### 3.40. Areas of High Radioactivity

Populations in other regions of the world exist in fields of much higher background radiation than the United States. These high radiation levels are similar to or above levels of exposure to workers in the nuclear industry.<sup>61</sup> Some families of these local populations have resided in these regions for several generations. Such regions are useful for the study of the effects of higher radiation exposures to humans by direct observation, without the need to extrapolate the effects of lower exposures of ionizing radiation based solely upon the effects of high exposures of ionizing radiation. Risk estimates based on extrapolations from exposure to high levels of ionizing radiation from such data as atomic bomb survivors and irradiated patients for cancer treatment, offer no accurate estimation as to the effects of exposures to low levels of ionizing radiation, taking into account the uncertainties due to typical linear, infralinear, and supralinear extrapolation models.<sup>62</sup>

### 3.41. Thermal Springs

Mineral springs are known to contain high concentrations of radium and radon. These springs are used as spas by many people in the world because of a belief in the alleged, beneficial health effects provided by the mineral springs. Visitors are encouraged not only to drink and bathe in these waters, but to sit in closed spas where they can inhale <sup>222</sup>Rn from surrounding rock.<sup>63</sup>

Location	<sup>226</sup> Ra (kBq/m <sup>3</sup> )		<sup>222</sup> Rn (kBq/m <sup>3</sup> )	
	Range	Energy Release (MeV)	Range	Energy Release (MeV)
Badgestein: AUS	.04 – 4.9	304. x 10 <sup>9</sup> – 37. x 10 <sup>12</sup>	20 – 4500	3. x 10 <sup>12</sup> – 655. x 10 <sup>12</sup>
Tuwa: IND	.4 – .9	3. x 10 <sup>12</sup> – 7. x 10 <sup>12</sup>	4 – 40	582. x 10 <sup>9</sup> – 6. x 10 <sup>12</sup>

TABLE 3.42: RELEASES IONIZING RADIATION FROM <sup>226</sup>RA AND <sup>222</sup>RN IN HOT SPRINGS AT 50 YEARS<sup>64</sup>

<sup>61</sup> Wei Luxin, "High Background Radiation Area – An Important Source of Exploring the Health Effects of Low Dose Ionizing Radiation," High Levels of Natural Radiation 1996: Radiation Dose and Health Effects, (Amsterdam: Elsevier Science B. V., 1997) 1.

<sup>62</sup> A. R. Oliveira, et. al., "Brazilian Research in Areas of High Natural Radioactivity," High Levels of Natural Radiation 1996: Radiation Dose and Health Effects, (Amsterdam: Elsevier Science B. V., 1997) 120.

<sup>63</sup> Eisenbud, 191.

<sup>64</sup> M. Sohrabi, "World High Level Natural Radiation and Radon Prone Areas with Special Regard to Dwellings," High Levels of Natural Radiation 1996: Radiation Dose and Health Effects, (Amsterdam: Elsevier Science B. V., 1997) 65.

### 3.42. Brazil

Some areas in Brazil exhibit high background radiation fields. In the Guarapari and Meaiepe regions, the occurrence of heavy minerals such as monazite, ilmenite, zircon, and rutile is due to heavy weathering and the decomposition of the massifs located parallel to the coastline.<sup>65</sup> After a reduction in size, minerals were carried away as alluvia by the rivers running through the region to the ocean. Subsequently, the minerals were stratified and deposited in river beds and on sea floors. The result is the formation of long strips of deposits of radioactive materials, intercalated with inactive layers of ordinary sand.<sup>66</sup> Monazite sand deposits can be found in the states of Espirito Santos and Rio de Janeiro. During the ten years under analysis (1985-1994) of which the first five were based on medical records, no indication was noted of any changes in the nosocomial profile, nor any alteration in cancer incidence, whose occurrence in the region is extremely low.<sup>67</sup>

Locality	<sup>226</sup> Ra (pCi/g ash)		Energy Release (MeV)	
Controls	.006 – .123	.027	2. x 10 <sup>6</sup> – 35. x 10 <sup>6</sup>	8. x 10 <sup>6</sup>
Guarapari	.006 – .104	.036	2. x 10 <sup>6</sup> – 29. x 10 <sup>6</sup>	10. x 10 <sup>6</sup>
Meaiepe	.006 – .077	.026	2. x 10 <sup>6</sup> – 22. x 10 <sup>6</sup>	7. x 10 <sup>6</sup>
Araxá	.006 – .204	.063	2. x 10 <sup>6</sup> – 57. x 10 <sup>6</sup>	18. x 10 <sup>6</sup>
Tapira	.008 – .174	.060	2. x 10 <sup>6</sup> – 49. x 10 <sup>6</sup>	17. x 10 <sup>6</sup>

TABLE 3.43: RELEASES OF IONIZING RADIATION FROM DIETARY INTAKES IN REGIONS OF BRAZIL AT 50 YEARS<sup>68,69</sup>

<sup>65</sup> Oliveira, 120.

<sup>66</sup> *Ibid.*, 120.

<sup>67</sup> *Ibid.*, 124.

<sup>68</sup> E. Penna Franca, "Review of Brazilian Investigations in Areas of High Natural Radioactivity," *Simpósio*

*Internacional Sobre Areas De Elevada Radioatividade Natural*, (Rio de Janeiro, Brazil: Academia Brasileira de Ciencias, 1977) 33.

<sup>69</sup> These concentrations are based upon the <sup>226</sup>Ra content in teeth.

### 3.43. India

The 55 km long coastal strip in Kerala province and the 2.5 km strip in Tamil Nadu province have high external radiation levels because of the high thorium content of monazite sands in these areas. This area also supports a high density of human and plant populations. Exposure rates in air have been reported to be 3.5 to 70 times the representative world average.<sup>70</sup> As a result, indoor exposure rates are quite high because dwellings in this region do not provide any significant shielding effects.

The biological systems in these areas, including human inhabitants, are subjected to exposures to ionizing radiation from external sources caused by beta particles and gamma rays from natural thorium contained in monazite, and internal exposures following the uptake and resultant deposition of these radioactive materials in the biological systems, which includes the entry of radionuclides in the human body through ingestion and through inhalation.<sup>71</sup>

The daily per capita dietary intake of <sup>228</sup>Ra and <sup>40</sup>K has been estimated to be 162 pCi and 3551 pCi, respectively.<sup>72</sup> Although exposure levels are significant, the results of a demographic survey for epidemiological studies on chromosomal anomalies of human blood cells, and plants, have shown no conclusive statistically significant biological effects on the population of high radiation areas in India in comparison to control groups.<sup>73</sup>

Radionuclide	Activity	Energy Release (MeV)
<sup>228</sup> Ra	162 pCi	3. x 10 <sup>9</sup>
<sup>40</sup> K	3551 pCi	275. x 10 <sup>9</sup>
<b>Total</b>		<b>278. x 10<sup>9</sup></b>

TABLE 3.44: RELEASES OF IONIZING RADIATION FROM DIETARY INTAKES IN REGIONS OF INDIA AT 50 YEARS

<sup>70</sup> Bennett, 21.

<sup>71</sup> Oliveira, 81.

<sup>72</sup> A. R. Gopal-Ayengar, , et. al., "Current Status of Investigations on Biological Effects of High Background Radioactivity in the Monazite Bearing Areas of Kerala Coast in Southwest India," Simposio Internacional Sobre Areas De Elevada Radioatividade Natural, (Rio de Janeiro, Brazil: Academia Brasileira de Ciencias, 1977) 25.

<sup>73</sup> Sohrabi, 61.

### **3.50. Summary**

Contributions to the overall, ambient level of ionizing radiation originate from several different radionuclides. Adjustments have been made in the quantification of these radionuclides in order to normalize the many differences among them. This normalization has resulted in the characterization of these radionuclides based upon committed releases of ionizing radiation to the environment. Because the characterization was developed from this perspective, this large system of ionizing radiation can serve as control base to which more specified inventories of radionuclides can be directly compared.

Many epidemiological studies concerning the effects of ionizing radiation on populations that dwell in areas of high background ionizing radiation have been conducted for the past two decades. Because of the highly considerable variations that occur regarding actual levels of ionizing radiation present in the environment, as well as the many differences that exist between the radionuclides themselves, many difficulties can arise when formulating substantial comparisons among various environmental regions throughout the planet. One such measure of the effects of radionuclides, is the quantity of ion pairs that will be imparted on biological material. The adjustments that have been made to existing radioactive quantification can provide a basis for further epidemiological studies regarding the effects of ionizing radiation with respect to significant human exposures.

## **4.00. Application**

### ***4.10. Introduction***

Quantification of radioactivity based upon releases of ionizing energy has normalized differences attributable to distinct radionuclides. Section 3.00 presented a characterization of natural radionuclide inventories based upon total committed energy releases over time. A time period of 50 years was used for these characterizations in order to represent a significant fraction of a human lifetime. In addition, a further characterization evaluated committed energy releases from areas that are known to exhibit high levels of background radiation. Committed energy releases from these areas can be directly related to epidemiological studies of the biological effects of ionizing radiation that have been conducted over the last two decades. These characterizations can be applied as a control inventory to which more specified systems of radionuclides can be compared.

In Section 3.00, energy releases from natural sources were evaluated on a unit mass or unit volume basis. If the actual size of a specific system can be estimated, then the overall energy release to the environment can be ascertained. The total energy releases from each radionuclide then can be added together in order to present an overall quantity of ionizing radiation that will be released to the environment. The quantities of energy released from these specific systems can be compared to the releases of ionizing radiation result from the control inventory of existing, natural radionuclides.

## 4.20. Airborne Releases

Human activities contribute to large fluxes of gases and particles in the atmosphere.<sup>74</sup>

The initial quantities of radionuclides that are released from a specific airborne system can be adjusted to reflect the commitment of ionizing energy that will be released over a given time period.

## 4.21. Coal Power Plants

The most significant burden from coal plants to the environment involves atmospheric releases. Such releases include SO<sub>2</sub>, NO, NO<sub>x</sub>, and CO. Coal itself contains quantities of naturally occurring radionuclides, such as <sup>40</sup>K, <sup>232</sup>Th, and <sup>238</sup>U. The combustion of coal results in elevated concentrations of these radionuclides in ash and in gaseous residues. The release of these residues to the environment, either through a stack or from a waste storage areas, results in the distribution of these natural radionuclides from deep inside the earth, to surface locations, where they can enhance the ambient radiation field, resulting in higher population exposures.<sup>75</sup>

Combustion redistributes radionuclides trapped in the coal into different environments in the biosphere. When coal is mined, radionuclides are brought to the biosphere from the overburden as well as from the coal. When coal is burned, most of it fuses into ash. Fly ash and slag comprise the total volume of ash produced. Fly ash is very fine, and it is formed when ash is entrained by flue gases. Fly ash is collected in the stack, but some of this material escapes to the atmosphere. Ash that is too heavy to be transported by flue gases settles at the bottom of the boiler. Bottom ash also contains some incompletely burned organic matter.

Coal ash is not sequestered from the environment, as in the case of the current retention and ultimate geological disposal of nuclear power radioactive wastes. All coal is ultimately disposed in the biosphere. The total quantity of ionizing radiation that is released to the biosphere from this system therefore can be estimated based on the concentrations of radionuclides in the original coal. For the various radionuclides that are discharged to the environment, the total amount of ionizing radiation released can be calculated based upon the committed energy attributable to each radionuclide in the system.

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<sup>74</sup> Harold F. Hemond and Elizabeth J. Fechner, Chemical Fate and Transport in the Environment, (New York: Academic Press, 1994) 235.

<sup>75</sup> T. V. Ramachandran and U. C. Mishra, "Radioactivity Levels in Indian Coal and Some Technologically Enhanced Exposure to Natural Radiation Environment in India," Ecology, vol. 3 no. 6, November 1988, 9.

#### **4.22. Nuclear Power Plants**

Nuclear power plants also are a source of airborne releases to the environment. Gaseous radionuclides that are released to the atmosphere are produced as fission products. The NRC has developed a normalized model for a “reference reactor year” for environmental reports prepared for the construction permit stage of a light water cooled nuclear power reactor. These reports evaluate the contribution of the environmental effects of typical uranium fuel cycle activities to the environmental costs of licensing the nuclear power reactor.



Radionuclide	Range	Energy Release (MeV)
$^{238}\text{U}$	.3 ppm – 14.1 ppm	$990. \times 10^{15} - 45. \times 10^{18}$
$^{232}\text{Th}$	1.8 ppm – 22.7 ppm	$450. \times 10^{15} - 6. \times 10^{18}$
$^{226}\text{Ra}$	3.7 Bq/kg – 185 Bq/kg	$252. \times 10^{15} - 9. \times 10^{18}$
$^{40}\text{K}$	18.5 Bq/kg – 284 Bq/kg	$351. \times 10^{15} - 5. \times 10^{18}$
Total		$2. \times 10^{18} - 65. \times 10^{18}$
Natural Radon	200 – 2000 pCi/L	$8. \times 10^{18} - 85. \times 10^{18}$
Total Equivalent Background		$27. \times 10^{18}$

**TABLE 4.21: ATMOSPHERIC RELEASES OF IONIZING RADIATION FROM COAL POWER PLANTS AT 50 YEARS**

Radionuclide	Annual Activity (Ci)	Energy Release (MeV)
$^{226}\text{Ra}$	.02	$178. \times 10^9$
$^{14}\text{C}$	24	$7. \times 10^{12}$
$^3\text{H}$	18100	$208. \times 10^{12}$
$^{129}\text{I}$	1.3	$378. \times 10^9$
$^{131}\text{I}$	.83	$880. \times 10^6$
Total		$216. \times 10^{12}$
Natural Radon	200 – 2000 pCi/L	$8. \times 10^{18} - 85. \times 10^{18}$
Total Equivalent Background		$27. \times 10^{18}$

**TABLE 4.22: ATMOSPHERIC RELEASES OF IONIZING RADIATION FROM A NUCLEAR POWER PLANT AT 50 YEARS**

Both Table 4.21 and Table 4.22 apply the method of quantifying the energy commitment of a release of ionizing radiation from a distinct system of radionuclides. Because concentrations have been based on an energy release, each radionuclide can be added together, in order to quantify emissions, instead of identifying the concentration of each radionuclide within the plume. A representative value of 10 tons was used to estimate the activity and subsequent release of ionizing radiation from a coal power plant.<sup>76</sup>

The quantity of *Natural Radon* refers to the average range of  $^{222}\text{Rn}$  that is contained in outdoor air, as described in Section 3.35.1. The total release of ionizing radiation, attributable to  $^{222}\text{Rn}$ , is based upon an average density of air equivalent to 10 tons of coal released to the

<sup>76</sup> T. V. Ramachandran and U. C. Mishra, "Radiological Impact of Airborne Releases From Coal Fired Power Plants in India," *Ecology*, vol. 4 no. 3, August 1989, 23

atmosphere. The quantity of *Total Equivalent Background* is based upon the average concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ , and  $^{40}\text{K}$  that were characterized in Section 3.00. The ionizing radiation due to each radionuclide has been added together to present a total quantity of energy that would be released, equivalent to 10 tons of coal.

Because characterizations are based upon energy releases, direct comparisons can be formulated between each system of radionuclides. The airborne releases from coal combustion to the environment are within the same order of magnitude, but slightly higher, as average  $^{222}\text{Rn}$  concentrations in outdoor air and average radionuclide concentrations in the soil. The average airborne releases from a nuclear power plant are quite low in comparison to these other systems of radionuclides. The nuclear power industry is the most heavily regulated industry with respect to other common power producing industries.

The time period for which the committed release of ionizing radiation resulting from these emissions to the environment is taken to be 50 years, since the concern is directed to the effects of human health resulting from exposure to airborne emissions. Concerning long term environmental damage, a time period of 500 years could be applied in order to reflect the total environmental effects of ionizing energy from these sources of ionizing radiation.

### 4.30. New Jersey Aquifer System

A large majority of the potable water supplies in this country is derived from groundwater aquifer systems. Recently, the USGS published a report detailing the results of a six year study of the natural radioactivity and inorganic chemistry of groundwater in the Kirkwood-Cohansey aquifer system, located in Southern New Jersey. This aquifer system is the principal source of potable water in the area. The aquifer is predominantly unconfined. Results of routine monitoring of radioactivity in public drinking water supplies that tap the Kirkwood-Cohansey aquifer system indicated the presence of elevated concentrations of radium in the groundwater in this area.<sup>77</sup>

### 4.31. USGS Data

The sum of the concentrations of dissolved  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in samples from the aquifer system exceeded the EPA maximum contaminant level (MCL) of 5 pCi/L. The concentrations ranged from 1.1 pCi/L and 14.5 pCi/L, with a median concentration of 3.85 pCi/L, and the ratio of the concentration of  $^{226}\text{Ra}$  to the concentration of  $^{228}\text{Ra}$  in most samples was approximately 1:1, thus indicating that the aquifer matrix is not enriched in uranium relative to thorium.<sup>78</sup>

Radionuclide	Total Activity (1:1 ratio)	Energy Release (MeV)
$^{226}\text{Ra} + ^{228}\text{Ra}$	1.1 pCi/L	$21. \times 10^{18}$
$^{226}\text{Ra} + ^{228}\text{Ra}$	3.85 pCi/L	$74. \times 10^{18}$
$^{226}\text{Ra} + ^{228}\text{Ra}$	14.5 pCi/L	$256. \times 10^{18}$
EPA MCL ( $^{226}\text{Ra} + ^{228}\text{Ra}$ )	5 pCi/L	$96. \times 10^{18}$
Small Community Supplies $^{222}\text{Rn}$	$10^4$ pCi/L – $10^5$ pCi/L	$7. \times 10^{21} - 69. \times 10^{21}$
Average $^{222}\text{Rn}$ Content <sup>79</sup>	117 pCi/L – 287 pCi/L	$81. \times 10^{18} - 198. \times 10^{18}$
EPA MCL for $^{222}\text{Rn}$	300 pCi/L	$256. \times 10^{18}$
Total Equivalent Background		$64. \times 10^{21}$

TABLE 4.31: RELEASES OF IONIZING RADIATION IN KIRKWOOD-COHANSEY AQUIFER SYSTEM AT 50 YEARS

<sup>77</sup> Jane Kozinski, et. al., "Natural Radioactivity in, and Inorganic Chemistry of, Groundwater in the Kirkwood-Cohansey Aquifer System, Southern New Jersey, 1983-89," United States Geological Survey: Water Resources Investigations Report, 92-4144, (1995): USGS Earth Science Information Center, Denver, CO, 3.

<sup>78</sup> New Jersey, 1.

<sup>79</sup> Eisenbud, 157.

The data presented in the USGS report added together the concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in order to formulate a total radium content in the groundwater. This addition is not entirely accurate because both of these radionuclides decay with different half lives and with different energies. Therefore, the energy releases shown in Figure 4.23 have been calculated to reflect the sum the proportional energy contributions of each radionuclide in the system, given a 1:1 ratio.

Table 4.31 presents the a characterization of a specific system of radionuclides in groundwater, with respect to an overall release of ionizing radiation. The total energy release was based upon an estimated aquifer size of  $127.8 \times 10^6 \text{ m}^3$ .<sup>80</sup> These data are compared to the average levels of  $^{222}\text{Rn}$  that are found in potable water supplies in the country. Average  $^{222}\text{Rn}$  concentrations were previously characterized in Section 3.00. The total equivalent background energy release was derived from the same data that was used in Figure 4.22. The ionizing energy released from both of these systems was based upon total releases of ionizing radiation, equivalent to the estimated aquifer size of  $127.8 \times 10^6 \text{ m}^3$ .

Comparisons between these systems of radionuclides can be formulated based upon the common releases of ionizing radiation. The releases of ionizing radiation due to the radium in New Jersey fall within the same order of magnitude as average levels of  $^{222}\text{Rn}$  in potable water supplies. The highest range of radium release is nearly equivalent to the EPA MCL for  $^{222}\text{Rn}$ . Both systems release quantities of ionizing radiation that are less than equivalent background releases based upon average, natural radionuclide inventories. These comparisons can be further characterized to aid in the pathway analyses of the relationship of radium bodily uptake to calcium in the bone with respect to radon uptake to the lungs.

The radium content in the groundwater in New Jersey is considered to be elevated because the samples have exceeded the EPA MCL of 5 pCi/L. Other areas of the world exhibit an elevated radium content as well. Two such regions are Badgestein, Austria, and Tuwa, India, in which local populations have used these springs for alleged, beneficial health effects.

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<sup>80</sup> This volume was estimated by direct observation from maps and data provided on pages 13-16 of the USGS report.

Location: Radionuclide	Activity	Energy
Badgestein, AUS: $^{226}\text{Ra}$	1.08 pCi/L – 132.3 pCi/L	$39. \times 10^{18} - 5. \times 10^{21}$
Tuwa, IND: $^{226}\text{Ra}$	10.8 pCi/L – 24.3 pCi/L	$383. \times 10^{18} - 895. \times 10^{18}$
Badgestein, AUS: $^{222}\text{Rn}$	540 pCi/L – 121500 pCi/L	$383. \times 10^{18} - 84. \times 10^{21}$
Tuwa, IND: $^{222}\text{Rn}$	108 pCi/L – 1080 pCi/L	$74. \times 10^{18} - 767. \times 10^{18}$
Badgestein: Total $^{226}\text{Ra} + ^{222}\text{Rn}$		$422. \times 10^{18} - 89. \times 10^{21}$
Tuwa: Total $^{226}\text{Ra} + ^{222}\text{Rn}$		$457. \times 10^{18} - 2. \times 10^{21}$

**TABLE 4.24: RELEASES OF IONIZING RADIATION FROM THERMAL SPRINGS AT 50 YEARS**

Figure 4.24 presents the energy releases from concentrations of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  in mineral springs in Austria and India, equivalent to the size of New Jersey aquifer system. These springs are used as spas by many people. Pathways of exposure to these systems of radionuclides are primarily inhalation of  $^{222}\text{Rn}$  ingestion of both  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$ . The committed release of ionizing radiation from these two systems of radionuclides clearly exceed those from the New Jersey aquifer system. In addition, both the committed releases of ionizing radiation based on the each EPA MCL falls within the average ranges of energy releases in both Austria and India. Data from areas such as mineral springs have been used in many epidemiological studies of elevated exposures to ionizing radiation throughout the world, in order to determine the biological effects of these higher levels of ionizing radiation on local populations. The results of these studies could be applied to similar areas of elevated concentrations in this country with respect to both the effects of elevated concentrations on local populations and the formulation of regulatory limits.

#### **4.40. High Level Radioactive Waste**

One quarter to one third of the total fuel loading in a nuclear power reactor is changed approximately every 12 to 18 months. This fuel is considered “spent,” as it is no longer efficient for producing effective levels of fission for the production thermal energy. Most of the ionizing radiation emanating from spent fuel originates from fission products. Spent fuel in the United States is stored on site in pools. As of December 1998, the United States had accumulated approximately 38500 metric tons of spent nuclear fuel.<sup>81</sup>

#### **4.41. Yucca Mountain**

As a result of the passage of the Nuclear Waste Policy Act (1982), the Department of Energy (DOE) has assumed responsibility for the establishment and the operation of an underground disposal facility for spent nuclear fuel and other high level radioactive waste. Congress passed amendments to this act in 1987, essentially directing DOE to focus solely on a repository at Yucca Mountain, Nevada. For over the past 15 years, DOE has been studying the site at Yucca Mountain in order to determine whether it is a suitable place at which to build a geologic repository for this type of waste. A repository would be built 1000 feet below the surface and 1000 feet above the water table.<sup>82</sup> Groundwater is isolated in a closed regional basin that does not flow into any rivers that reach the ocean.<sup>83</sup>

Because of the eventual position of the repository, and because all of the wastes that will be stored there are solids, the only pathway that would result in a release of radionuclide quantities to the environment is through the groundwater system. The rock mass at Yucca Mountain is composed of volcanic rock that is fractured as a result of contraction during cooling of the original molten rock. Water flowing through these fractures will move more rapidly than water flowing through the soil matrix. The overall vadose zone flow system is very heterogeneous.<sup>84</sup> During the thousands of years that will pass before water could infiltrate a degraded waste package, most radionuclides will have decayed to a near stable state. For the release of remaining radionuclides to occur, they must be dissolved in water. Only long lived water soluble radionuclides such as <sup>99</sup>Tc, <sup>129</sup>I, and <sup>237</sup>Np could be released. Both <sup>242</sup>Pu and <sup>239</sup>Pu also could be released, but these radionuclides most likely will adsorb to colloids in the vadose zone, thus slowing transport significantly. These radionuclides will have to travel 1000 feet down to infiltrate the water table and nearly 12 miles to reach a point at which uptake from a well which would constitute an exposure pathway.

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<sup>81</sup> United States Department of Energy, Office of Civilian Waste Management, Yucca Mountain Site Characterization Office, Viability Assessment of a Repository at Yucca Mountain: Overview, December 1998, <http://www.ymp.gov/vadoc/overview/p1.pdf>, 4

<sup>82</sup> “*Environmental Impact Statement*,” <http://www.ymp.gov/timeline/va/vanew.htm>.

<sup>83</sup> Viability Assessment: Overview, 2.

<sup>84</sup> United States Department of Energy, Office of Civilian Waste Management, Yucca Mountain Site Characterization Office, Viability Assessment of a Repository at Yucca Mountain: Volume 3: Total System Performance Assessment, December 1998, <http://www.ymp.gov/vadoc/va3/p3.pdf>, O-7.

The appropriate objective for the assessment of this system of radionuclides is the minimization of the potential exposure to ionizing radiation of the local population living downgradient from the site. Evaluations of energy releases that could cause such exposures can be formulated and eventually compared to the effects of similar exposure levels already existing in the environment.

#### **4.41.1. Viability Assessment**

In December 1998, the Yucca Mountain Project published the *Viability Assessment of a Repository at Yucca Mountain* (VA). The report concluded that work should continue toward the formulation of a final decision in 2001 concerning the development of a geological repository at this site. The performance assessment for this repository design indicates that for 10000 years after the repository is closed, populations dwelling in the vicinity of this area would receive little increase in radiation exposure. In addition, after approximately 300000 years, populations dwelling within 12 miles of Yucca Mountain could receive additional exposures that are comparable to present day exposures to natural background radiation.<sup>85</sup> The nearest population that could be exposed to releases from the repository is assumed to be 12 miles from the site boundary.

#### **4.41.2 Spent Fuel Inventory**

Some sections in the VA present the total inventory of spent fuel strictly in terms of a cumulative activity. The manner in which this system of radionuclides is classified is not entirely accurate. Aggregate inventories of radionuclides cannot be represented properly though an addition of individual activities because usage of the unit of activity itself is specific to each radionuclide. The addition of 2 Ci of <sup>237</sup>Np with 1 Ci of <sup>99</sup>Tc will not result in a total radionuclide inventory of 3 Ci. If a total representation of the quantities of radionuclides in a system is to be characterized, then the inventory can be adjusted to reflect the committed releases of ionizing radiation, prior to the addition of any radionuclide quantities.

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<sup>85</sup> "Environmental Impact Statement," <http://www.ymp.gov/timeline/va/vanew.htm>.

Radionuclide	Quantity (Ci/pkg)	50 Years	10 <sup>5</sup> Years	10 <sup>6</sup> Years
<sup>237</sup> Np	11.4	3. x 10 <sup>24</sup>	4. x 10 <sup>27</sup>	20. x 10 <sup>27</sup>
<sup>99</sup> Tc	118	480. x 10 <sup>21</sup>	810. x 10 <sup>24</sup>	3. x 10 <sup>27</sup>
<sup>239</sup> Pu	3050	300. x 10 <sup>24</sup>	420. x 10 <sup>27</sup>	3. x 10 <sup>30</sup>
<sup>242</sup> Pu	17	5. x 10 <sup>24</sup>	7. x 10 <sup>27</sup>	30. x 10 <sup>27</sup>
<b>Total</b>		<b>300. x 10<sup>24</sup></b>	<b>420. x 10<sup>27</sup></b>	<b>3. x 10<sup>30</sup></b>

**TABLE 4.51: COMMITTED RELEASES OF IONIZING RADIATION FROM RADIONUCLIDES IN SPENT NUCLEAR FUEL<sup>86</sup>**

Table 4.51 applies the method of quantifying a specific system of radionuclides according to a committed release of ionizing radiation. This adjustment from singular activities to committed releases of ionizing radiation allows for the addition of each radionuclide within the system. This adjustment can provide a representation of the full amount of ionizing radiation that the system could release to the biosphere. The radionuclides in this system are those quantities which most likely will be released to the biosphere at some time after the closure of the repository, as a result of waste package degradation. These adjustments were based upon an estimated total of 7760 commercial spent nuclear waste packages to be emplaced within the repository, and a maximum 3% cladding failure.<sup>87</sup> Thus, the releases of ionizing radiation from the radionuclides in the spent fuel inventory presented in Table 4.51 represent the quantity of radionuclides that will be exposed to the environment in the repository and will be available for transport and for eventual release to the biosphere. Presumably, the VA is based upon the assumption that only a fraction of these quantities will actually be released.

#### **4.41.3. Modeling Parameters**

Processes such as advection, sorption, diffusion, dispersion, and decay will affect the transport of radionuclides, after waste package degradation has occurred. The consideration of decay chains causes transport modeling to be more complex because of the formation of daughter radionuclides. The creation of progeny could be characterized in a complex solution as a distributed source term throughout the zone of transport; however, the vadose zone transport model used in the VA does not directly consider the ingrowth of progeny.

Transport through the saturated zone can affect the release of ionizing radiation to the biosphere by delaying the release of the radionuclides and by diluting concentrations during migration. Radionuclides are expected to move through the unsaturated zone below the repository and enter the saturated zone in a well defined plume. Three dimensional modeling

<sup>86</sup> VA: Volume 3, 3-96.

<sup>87</sup> VA: Volume 3, 4-60.



was used only to determine flow paths in the saturated zone. One dimensional transport modeling was used, based on the flow paths, to determine concentration breakthrough curves for unit releases of radionuclides.<sup>88</sup> Ingrowth of progeny also was not considered for transport in the saturated zone.

#### **4.41.4. Fractional Releases**

Another manner in which to examine the releases of ionizing radiation from the spent fuel inventories to the biosphere is through an assessment of the cumulative quantities of radionuclide releases in the saturated zone, at the 12 mile point at which exposure pathways should be considered.

Releases from individual radionuclides to the biosphere are given according to a cumulative fractional release at the 12 mile boundary. This cumulative fractional release is defined by the cumulative activity that has traveled past the 12 mile boundary divided by the initial amount of radioactivity emplaced in the repository for the radionuclide.<sup>89</sup>

<b>Radionuclide</b>	<b>Activity (Ci)</b>	<b>Energy Release</b>
<sup>99</sup> Tc	275	5. x 10 <sup>21</sup>
<sup>237</sup> Np	27.	29. x 10 <sup>21</sup>
<sup>242</sup> Pu	2.	3. x 10 <sup>21</sup>
Total		37. x 10 <sup>21</sup>
Total Equivalent Background		Table 4.53.1

**TABLE 4.53: RELEASES OF RADIONUCLIDES FROM THE SATURATED ZONE AT 300000 YEARS**

<sup>88</sup> VA: Volume 3, 3-139.

<sup>89</sup> VA: Volume 3, 4-61.

The data presented in Table 4.53 represents the estimated, 50 year, committed releases of ionizing radiation from the saturated zone to the biosphere that could occur at 300000 years following repository closure, at the 12 mile boundary.<sup>90</sup> A premise concluded in the VA is that is that releases from the repository could produce additional exposures that are comparable to current exposures from natural background radiation. The quantification of these radionuclides has been adjusted to reflect the total release of ionizing radiation at the 300000 year period. These data caqn be compared to the present natural radionuclide inventory that was characterized in Section 3.00. The flow through the saturated zone is very complex. The saturated zone itself is fairly large. Based on data in the VA, concentrations of these radionuclides are fairly consistent throughout transport in the saturated zone. These radionuclides presumably will be released to the biosphere directly from the water table as a result of pumping at a well, whether for usage in an irrigation system or for usage as a potable water supply. Therefore, these releases most likely will occur over a small fraction of the groundwater system.

Fraction of Groundwater System	Equivalent Background Radiation (MeV)
.01	$35. \times 10^{24}$
.001	$4. \times 10^{24}$
.0001	$350. \times 10^{21}$
.00001	$35. \times 10^{21}$
.000001	$4. \times 10^{21}$

**TABLE 4.53.1: EQUIVALENT BACKGROUND RADIATION BASED ON FRACTIONS OF THE GROUNDWATER SYSTEM<sup>91</sup>**

Although these estimates are uncertain, the analysis upon which they are based is reasonable, given the data and maps provided in the VA. In addition, this analysis presents yet another opportunity in which quantities of radionuclides in a specific system can be adjusted to provide for direct comparisons with another system of radionuclides. The estimates somewhat agree with the premise in the VA that energy releases from the radionuclides in the repository will provide additional exposures that are comparable to present day exposures from natural sources of ionizing radiation. A further characterization of the area at which the radionuclides will be released to the biosphere, with regard to the inventory of natural radionuclides that are specific to the region, as well as the pathways from which all of these releases could occur, will most likely will provide for better estimates to which all these quantities of radionuclides can be compared.

<sup>90</sup> Size of the saturated zone used in the model was estimated to be 20 km x 36 m x 950 m, divided into 17 different units. (VA: Volume 3, 3-135.)

<sup>91</sup> Estimations based upon data and maps provided in the VA.

#### **4.50. Summary**

Section 3.00 applied the method of adjusting quantities of radionuclides to reflect a committed release of ionizing radiation over a specific time period. A time period of 50 years was used in order to characterize a significant fraction of a human lifetime. The quantities of radionuclides that were adjusted in Section 3.00 involved sources of ionizing radiation that are commonly found in nature. Because these radionuclides have been adjusted on the common basis pertaining to the release of ionizing radiation, they can serve as a control inventory to which more specific systems of radionuclides can be directly compared.

Section 4.00 presented specific systems of radionuclides. Such systems include the airborne releases of radionuclides from both a coal power plant and a nuclear power plant, quantities of radionuclides in a groundwater system, and a discussion regarding the extensive modeling and characterization that is involved at the Yucca Mountain Project. The quantification of each system was adjusted to represent each radionuclide within the inventory as a source of ionizing radiation. In addition, a further analysis attempted to estimate the full extent of each system, in order to provide a total, overall release of ionizing radiation from the environment from the system. The releases of ionizing radiation attributable to each system then could be compared to releases of ionizing radiation from natural sources of ionizing radiation, of an equivalent extent.

Several conclusions have been formulated regarding the releases of ionizing radiation from a specific system of radionuclides. The releases of ionizing radiation of radionuclides from a coal power plant exceeded similar airborne releases from a nuclear power plant. Releases of ionizing radiation resulting from elevated concentrations of radium in the groundwater system in Southern New Jersey fell below the average releases of ionizing radiation with respect to radon in potable drinking water supplies, as well as the releases of ionizing radiation from average concentrations of natural radionuclides. An analysis of the possible future releases of ionizing radiation from radionuclides in spent nuclear fuel to the environment at Yucca Mountain, from the perspective of the committed energy release adjustment, remained fairly consistent with the premise that future radionuclide releases would result in additional exposures to ionizing radiation that are comparable to present day exposures from natural radionuclides. These analyses can be applied to other specific systems of radionuclides in the same manner, perhaps incorporating this adjustment to regions in the world known to exhibit high levels of background radiation with epidemiological evaluations.

The adjustment of radionuclide quantification to the perspective of a release of ionizing radiation has normalized the differences that inherently exist among all radionuclides. The analyses conducted in this manner are by no means a substitute for existing methods of environmental modeling or radiological studies. These types of analyses, however, can be incorporated into existing characterizations in order to present a more clear and a more accurate assessment of the effects of ionizing radiation, whether to the environment as a whole or to the human body.

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